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Resistance controllability and variability improvement in a TaO_x-based resistive memory for multilevel storage application

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In order to obtain reliable multilevel cell (MLC) characteristics, resistance controllability between the different resistance levels is required especially in resistive random access memory (RRAM), which is prone to resistance variability mainly due to its intrinsic random nature of defect generation and filament formation. In this study, we have thoroughly investigated the multilevel resistance variability in a TaO_x-based nanoscale (<30 nm) RRAM operated in MLC mode. It is found that the resistance variability not only depends on the conductive filament size but also is a strong function of oxygen vacancy concentration in it. Based on the gained insights through experimental observations and simulation, it is suggested that forming thinner but denser conductive filament may greatly improve the temporal resistance variability even at low operation current despite the inherent stochastic nature of resistance switching process. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4922446]

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

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In RRAM, MLC characteristics can be obtained by various methods such as by varying the switching current (or compliance current), where different levels of low resistance state (LRS) can be obtained due to change in the total number of constituent defects of the conductive filament (CF) or 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.^{10,11} MLC behavior by varying the program/ 51 erase pulse width is also reported.¹¹ 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.¹² 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

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

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

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FIG. 1. (a) Forming and resistive switching behavior of fabricated 30 nm sized TaO_x -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_C. Both decrease with increasing I_C. (e) Evolution of LRS variability (σ/μ) and reset current (I_{reset}) with I_C.

thick SiO₂ layer. Tantalum oxide (TaO_x) switching layer of ~10 nm was deposited by reactive sputtering using Ta metal target and Ar+O₂ 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_x switching layer. For electrical measurements, voltage was applied on TE and BE was connected to the ground.

Multilevel cell operation and resistance variability anal-92 ysis of fabricated 30 nm size RRAM device is presented in 93 Fig. 1. First, the device was formed by applying a negative 94 95 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 96 device exhibits resistive switching behavior and MLC char-97 acteristics by varying the switching current, as shown in 98 Figs. 1(a) and 1(b). Multiple cycles for each level have been 99 shown to ensure the stable resistance switching. Three differ-100 ent levels of LRS were obtained with same HRS, which can 101 be used in 2-bit per cell storage. In addition, it was observed 102 that the LRS current-voltage (I-V) gradually shifted from an 103 ohmic to a nonlinear behavior as current compliance $(I_{\rm C})$ 104 was decreased down to 50 μ A (Fig. 1(c)) due to decreased 105 number of vacancy defects in the CF leading to the change 106 in the current conduction mechanism. Simultaneously, the resistance margin between the resistance levels is reduced 108 because of cycle-to-cycle resistance fluctuations. The resist-109 ance distribution of 50 consecutive cycles for each compli-110 ance is shown in Fig. 1(d), where the highest variability of 111 14.6% is observed for I_C of 50 μ A. The read voltage was 112 0.25 V. Furthermore, the resistance of LRS (R_{LRS}) decreased 113 with increasing I_C [Fig. 1(d)]. The slope of R_{LRS} vs I_C was 114 found to be -0.9, which is close to the reported value of 115 $-1.^{12}$ On the other hand, maximum reset current (I_{reset}) 116 increases linearly and coefficient of variation (σ/μ) or 117 variability of R_{LRS} decreases with increasing I_C, as shown in 118 Fig. 1(e). The decrease in the variability is due to the 119 increase in the number of oxygen vacancy defects in the CF 120 with increasing I_C. Considering the dependence of LRS I-V 121 nonlinearity on I_C and variability behavior, it can be said that 122 by increasing I_C, both CF size and oxygen vacancy defect 123 density in it may increase. In other words, R_{LRS} depends on 124 filament size and its electrical conductivity. 125

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 128 oxygen vacancy concentration (n_{Vo}) falling under a thresh-129 old value (n_{TAC}) , which is the minimum concentration of 130 oxygen vacancies below which trap assisted conduction 131 (TAC) dominates and I-V becomes nonlinear, while above 132 this concentration conduction is linear (ohmic).^{15–18} This 133 can be accounted by the following equation: 134

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

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

where σ_e is the electrical conductivity, E_a is the activation 135 energy, $\beta = 1 \times 10^{-23}$ S m⁻¹, k = Boltzmann constant, 136 T = absolute temperature, and $E_{a0} = 0.4$ eV. This approach 137 has already been employed to describe resistance switching 138 phenomena or early data-retention failure in RRAM 139 employing either HfO_x^{15,16} or TaO_x.^{17,18} The corresponding 140 plot of σ_e as a function of n_{Vo} is shown in Fig. 2. In the 141 previous studies,^{15–18} a piecewise linear and decreasing 142 function *f* [Eq. (2)] was employed to account for the 143 decrease of conduction activation energy at increasing 144 defect concentrations which, in turns, increases σ_e in the CF 145 region. Although a proper theoretical justification is still 146



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_{Vo} .

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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} .

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

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

where t_{ox} is the oxide thickness (= 10 nm) and r_{CF} is the conductive filament radius. Figure 3(b) shows the R_{LRS} isovalue curves of all the resistance levels as a function of CF radius and oxygen vacancy concentration (n_{Vo}). The n_{Vo} equal to n_{TAC} is also shown by a dashed line. The dependence of 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 μ A 181 (respectively, 200 μ 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

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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 μ A. Monte-Carlo calculations have been 190 performed to reproduce the experimental LRS resistance dis- 191 tribution 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%) variation 196 in n_{V_0} can result in a large (14.6%) variability in the resist- 197 ance for 50 μ A I_C and a 5.9% (smaller) resistance variation 198 for 200 μ 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 $Ic = 200 \ \mu A$) and 16.7 meV (for $Ic = 50 \ \mu A$) [Fig. 4(e)]. E_a 204 values are in the same order of magnitude than those already 205 reported by Kim et al.^{17,18} 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.

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FIG. 5. (a) R_{LRS} isoline for $I_C = 50 \,\mu$ A. Shaded regions correspond to trapassisted (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 $n_{vo}/$ $n_{\rm TAC} > 1$ allows for smaller relative variation of $R_{\rm LRS}.$

minimize the resistance variations even at low I_C. It is possi-207 ble to improve the variations if we can shift to operate in the 208 linear region instead of nonlinear region, as shown in Fig. 209 210 5(a). This can be achieved by generating smaller size fila-211 ment with higher defect concentration. Calculations show that a tighter distribution with 8% variability can be achieved 212 213 even at low I_C of 50 μ 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 0.35 214 215 [Fig. 5(b)]. Therefore, forming denser but thinner filament may remarkably improve the resistance fluctuation even at 216 low operation current. Various techniques such as two step 217 forming, material engineering, and different forming condi-218 tions can be exploited to generate different defect density 219 filament.6 220

In this work, multilevel resistance variability was inves-221 222 tigated on a nanoscale TaO_x-based RRAM operating in MLC mode by means of a theoretical approach assuming 223 that non-linearity observed at low I_C is related to trap 224 assisted conduction resulting from low concentration of oxy-225 gen vacancy. It was found that the resistance variability not 226 only depends on the conductive filament size but also is a 227 strong function of oxygen vacancy concentration and fluctua-228 tion, which is supported by Monte-Carlo calculations of LRS 229 distributions at various compliance currents. Based on this 230 231 study, it is suggested that LRS variability can be improved 281

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even at low I_C by forming denser CF in terms of oxygen va- 232 cancy by means of material engineering or dedicated form- 233 ing techniques. 234

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¹H. S. Philip Wong, H.-Y. Lee, S. Yu, Y.-S. Chen, Y. Wu, P.-S. Chen, B. 239 Lee, F. T. Chen, and M.-J. Tsai, Proc. IEEE 100, 1951 (2012). 240

- ²F. Pan, S. Gao, C. Chen, C. Song, and F. Zeng, Mater. Sci. Eng., R 83, 1 241 242 (2014).
- ³A. Prakash, D. Jana, and S. Maikap, Nanoscale Res. Lett. 8, 418 (2013). 243 ⁴J. J. Yang, M. X. Zhang, J. P. Strachan, F. Miao, M. D. Pickett, R. D. 244 Kelley, G. Medeiros-Ribeiro, and R. S. Williams, Appl. Phys. Lett. 97, 245 232102 (2010). 246
- ⁵M. Lee, C. Lee, D. Lee, S. R. Lee, M. Chang, J. H. Hur, Y. B. Kim, C. J. 247 248 Kim, D. H. Seo, S. Seo, U. I. Chung, I. K. Yoo, and K. Kim, Nat. Mater. 249 10, 625 (2011).
- ⁶T. Ninomiya, Z. Wei, S. Muraoka, R. Yasuhara, K. Katayama, and T. 250 251 Takagi, IEEE Trans. Electron Devices 60, 1384 (2013).
- ⁷B. Govoreanu, G. S. Kar, Y.-Y. Chen, V. Paraschiv, S. Kubicek, A. 252 253 Fantini, I. P. Radu, L. Goux, S. Clima, R. Degraeve, N. Jossart, O. 254 Richard, T. Vandeweyer, K. Seo, P. Hendrickx, G. Pourtois, H. Bender, L. 255 Altimime, D. J. Wouters, J. A. Kittl, and M. Jurczak, IEDM Tech. Dig. 256
- 2011.729 ⁸C. H. Ho, C.-L. Hsu, C.-C. Chen, J.-T. Liu, C.-S. Wu, C.-C. Huang, C. Hu, 257 258 and F.-L. Yang, IEDM Tech. Dig. 2010, 436.
- ⁹S.-G. Park, M. K. Yang, H. Ju, D.-J. Seong, J. M. Lee, E. Kim, S. Jung, L. 259 260 Zhang, Y. C. Shin, I.-G. Baek, J. Choi, H.-K. Kang, and C. Chung, IEDM 261 Tech. Dig. 2012, 501.
- ¹⁰M.-C. Wu, W.-Y. Jang, C.-H. Lin, and T.-Y. Tseng, Semicond. Sci. 262 263 Technol. 27, 065010 (2012).
- ¹¹S. Yu, Y. Wu, and H.-S. P. Wong, Appl. Phys. Lett. 98, 103514 (2011). 264 ¹²S. Ambrogio, S. Balatti, A. Cubeta, A. Calderoni, N. Ramaswamy, and D. 265 Ielmini, IEEE Trans. Electron Devices 61, 2912 (2014). 266
- ¹³S. Ambrogio, S. Balatti, A. Cubeta, A. Calderoni, N. Ramaswamy, and D. 267 Ielmini1, IEDM Tech. Dig. 2013, 782. 268
- ¹⁴A. Fantini, L. Goux, R. Degraeve, D. J. Wouters, N. Raghavan, G. Kar, A. 269 270 Belmonte, Y.-Y. Chen, B. Govoreanu, and M. Jurczak, in Proceedings of 271 the IEEE International Memory Workshop, 2013, p. 30. ¹⁵S. Larentis, F. Nardi, S. Balatti, D. C. Gilmer, and D. Ielmini, IEEE Trans. 272
- Electron Devices 59, 2468 (2012). 273 ¹⁶T. Cabout, E. Vianello, E. Jalaguier, H. Grampeix, G. Molas, P. Blaise, O. 274
- 275 Cueto, M. Guillermet, J. F. Nodin, L. Perniola, S. Blonkowski, S. Jeannot, S. Denorme, P. Candelier, M. Bocquet, and C. Muller, in Proceedings of 276 277 the IEEE International Memory Workshop, 2014, p. 22. 278
- ¹⁷S. Kim, S. Choi, and W. Lu, ACS Nano 8, 2369 (2014).
- ¹⁸S. Kim, S.-J. Kim, K. M. Kim, S. R. Lee, M. Chang, E. Cho, Y.-B. Kim, 279 C. J. Kim, U.-I. Chung, and I.-K. Yoo, Sci. Rep. 3, 1680 (2013). 280

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