Role of Oxygen Defects in Conductive-Filament Formation in Y_2O_3 -Based Analog RRAM Devices as Revealed by Fluctuation Spectroscopy

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Low-frequency noise in Y_2O_3 -based resistive random-access memory devices with analog switching is studied at intermediate resistive states and as a function of dc cycling. A universal $1/f^\alpha$ -type behavior is found, with a frequency exponent of $\alpha \approx 1.2$ that is independent of the applied reset voltage or the device resistance and is attributed to the intrinsic abundance of oxygen vacancies unique to the structure of yttria. Remarkably, the noise magnitude in the high resistive state systematically decreases through dc training. This effect is attributed to the stabilization of the conductive filament via the consumption of oxygen vacancies, thus reducing the number of active fluctuators in the vicinity of the filament.

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I. INTRODUCTION

The bottleneck in the advancement of modern computing is not the calculating power itself but the way in which data are stored and managed: 90% of the total energy is consumed by data transport and storage [1]. Additionally, with the downscaling of electronic devices, the current baseline memory technology is approaching its limits in size and performance [2]. These are strong driving forces behind the extensive research to find new solutions in both computing methods and data storage. Resistive random-access memory (RRAM) is one of the most promising candidates amongst emerging nonvolatile memories [3] because of its high performance [4], good retention properties, high endurance [5], excellent scaling potential [6], and insensitivity to ionizing radiation [7]. Data storage in RRAM is based on the resistive-switching effect, where the resistance of the memory cell is changed between a high resistive state (HRS) and a low resistive state (LRS). The

transition from the HRS to the LRS is denoted as “set” and is related to the formation of a defect-rich path, commonly referred to as the conductive filament (CF), while switching from the LRS to the HRS, i.e., the “reset” process is explained by the rupture of the CF. If these transitions are analog, resistive switching can be exploited for establishing multilevel data storage, as well as electronic synapses for neuromorphic computing applications [8]. In devices based on the valence-change mechanism (VCM), oxygen engineering has been proven to be effective in achieving gradual switching dynamics and in lowering the forming and operation voltages via the introduction of an even distribution and high concentration of oxygen vacancies to the functional layer [9–12]. However, in transition-metal oxides, such as HfO_x , TaO_x or ZrO_x , which are often utilized as the switching medium, suboxide formation of various stoichiometries due to several possible valence states of the transition metal may lead to unreliability and device degradation [13,14]. For yttrium oxide, a promising candidate for nonvolatile memory [15–17] and neuromorphic applications [17–19], the only stable stoichiometry is Y_2O_3 , which crystallizes in the cubic C-type fluorite (bixbyite, sesquioxide) structure. YO, a metastable composition with an orthorhombic $Pnma$ structure, can also be viewed as oxygen-deficient cubic- Y_2O_3 and thus is easily embeddable into the stoichiometric yttria matrix [20].

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An intriguing feature of the bixbyite structure is that it has an intrinsically high defect density, as 25% of the anion sublattice sites remain unoccupied. The resulting so-called constitutional oxygen vacancies have been reported by several publications [20–22] to form nonintersecting vacancy chains or channels along preferred orientations of the crystal structure. These channels may assist filament growth, resulting in forming-free devices [15,17,20]. Thus yttria, being inherently rich in defects, is especially interesting for RRAM applications and is a great platform to investigate defect-induced electronic fluctuations.

In filamentary RRAM, read-current fluctuations can have a severe impact on device performance, as they can cause erratic data readout for memory applications and introduce unreliability to synaptic operations [23–28]. Conductance fluctuations can be caused by structural changes in the CF [29,30] and/or (de)trapping of electrons at dominant traps [31], namely random telegraph noise (RTN). The source of such fluctuations has been attributed to oxygen vacancies [25,32] and interstitials [24] existing in different charged states in HfO_{2-x} -based RRAMs. Low-frequency noise (LFN) associated with the aforementioned processes has long been known in electronic devices [33] and is often viewed as a reliability issue; however, it can also be used as a powerful nondestructive probing tool in analyzing the nature and physical origin of the fluctuations. In this study, LFN measurements are utilized in order to assess the performance of Y_2O_3 -based RRAM devices with analog switching characteristics in both multilevel and two-level operation modes and to gain insight into the underlying physical processes governing resistive switching.

II. EXPERIMENTAL

The devices under test (DUTs) consist of $\text{TiN}/\text{Y}_2\text{O}_3/\text{Pt}$, the fabrication of which is described in Ref. [17]. All electrical measurements are carried out at room temperature and ambient atmosphere. A Keithley 4200 semiconductor characterization system (SCS) is used to obtain the current-voltage (I - V) characteristics and to set the DUT to the desired resistance state for the noise measurements by applying a voltage bias to the Pt top electrode while keeping the TiN bottom electrode grounded and using the internal current compliance of the SCS to inhibit hard dielectric breakdown of the DUT. A JEOL JEM ARM-200F is used to obtain the scanning transmission electron microscopy (STEM) images. Low-frequency fluctuation spectroscopy measurements are carried out in a two-terminal dc configuration under constant voltage bias, which is supplied by a Keithley 2400 voltage source. The current fluctuations are amplified by a Keithley 428 current amplifier and the noise spectra are obtained by a Stanford Research 785 spectrum analyzer. For more details on the noise measurements, see Fig. S1 in the Supplemental Material [34]. The

current noise power spectral density (PSD), i.e., the mean random current squared per unit frequency band, is defined by [35]

$$S_I(f) = 2 \lim_{T \rightarrow \infty} \frac{1}{T} \left| \int_{-T/2}^{+T/2} dt e^{-i\omega t} \delta I(t) \right|^2, \quad (1)$$

where $\delta I(t)$ denotes the fluctuating part of the current signal measured over the time interval T and $\omega = 2\pi f$ is the angular frequency. Detailed information can be found in Refs. [35–37].

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the bright-field (BF) STEM overview image of the whole device stack and of the Y_2O_3 layer, respectively, revealing a polycrystalline structure. The high-resolution contrast enables the visualization of individual grains. Figure 1(c) shows the magnified high-resolution high-angle annular dark-field (HAADF) STEM image of the marked area in Fig. 1(b), with a multislice [38] simulation along the $(1\bar{1}0)$ direction. The C-type fluorite structure in which yttrium oxide crystallizes is schematically shown in Fig. 1(d), revealing the positions of yttrium atoms existing in two different oxygen environments [39], denoted here as type I and type II. All yttrium atoms are sixfold-coordinated with the type-I atoms having six oxygen neighbors at equivalent distances and the type II with three oxygen pairs at different distances

FIG. 1. (a) A BF-STEM image of the whole device stack. (b) A BF-STEM image of the Y_2O_3 thin film. The high-resolution contrast allows the identification of individual grains in the film. (c) A magnified HAADF-STEM view of the grain marked in (b); the inset shows the multislice [38] simulation along the $(1\bar{1}0)$ direction of Y_2O_3 . (d) The unit cell of the Y_2O_3 sesquioxide structure. (e) The position of oxygen vacancies arising from the environment of the two nonequivalent (type-I and type-II) yttrium lattice sites.

[40]. This specific structure results in vacant sites on the anion sublattice, marked by the green boxes in Fig. 1(e), totaling in a lattice-site occupation of 75%.

The low-frequency current noise characteristics are investigated as a function of the maximum applied voltage in the reset process, which is incremented from +2 V to +2.5 V in 0.1-V steps, denoted as $V_{\text{reset,max}}$ in Figs. 2(a)–2(e). To ensure the stability of the thus accessed different current levels, the devices are cycled five times with each $V_{\text{reset,max}}$ before the noise measurements are carried out with a readout voltage of -0.1 V. Representative current power spectral density curves normalized by the square of the average current are shown in Fig. 2(a) for one of the DUTs. For all measurements, a $1/f^\alpha$ -type frequency dependence is observed at lower frequencies, whereas for $f \gtrsim 10$ –20 Hz, the background noise of the experimental setup becomes dominant. $1/f$ noise in RRAM is usually attributed to charge-capture and -emission processes at multiple noncorrelated defect sites, whereas a $1/f^2$ frequency dependence, associated with the high-frequency tail of a Lorentzian spectrum, is the signature of a single fluctuator [26,27,41]. This also means that for a high defect density, mainly a $1/f$ -type frequency dependence is expected, with α close to 1. Deviations from “perfect” $1/f$ noise ($\alpha = 1$) can be explained by a nonconstant distribution of activation energies $D(E)$ of the individual two-level fluctuation processes [35]. In general, a superposition of $1/f$ and (even multiple) Lorentzian spectra is also possible and has been observed for other RRAM systems [27]. In Figs. 2(b)–2(d), the frequency exponent α , the normalized power spectral density at 1 Hz, and the resistance are plotted, respectively, as the function of $V_{\text{reset,max}}$ for four measured devices. At selected values of $V_{\text{reset,max}}$, the noise measurements are repeated in order to demonstrate reproducibility. The noise magnitude, the frequency exponent, and the DUT resistance show uniform characteristics for all measured devices, indicating a low device-to-device variation. Within the experimental error, the frequency exponent is found to be $\alpha \approx 1.2$ for all DUTs, independent of $V_{\text{reset,max}}$ or the device resistance, which means that the distribution of spectral weight is not affected by these parameters. Also, this $1/f$ -type behavior, with the frequency exponent so close to 1, indicates that the dominating source of fluctuations can be attributed to charge trapping and detrapping events at several noncorrelated traps, with a nearly constant distribution of activation energies of the individual fluctuators. The I - V characteristics for one of the devices are shown in Fig. 2(e). By incrementing $V_{\text{reset,max}}$, intermediate resistive states (IRSs) are accessed through the transition from the LRS to the HRS and the DUT resistance at these states increases from approximately 1 k Ω to approximately 100 k Ω . In terms of the CF structure, this transition can be understood with the decreasing cross section and the eventual breaking of the filament, as shown schematically in Fig. 2(f). Starting

FIG. 2. Low-frequency noise characteristics of the HRS obtained with different values of $V_{\text{reset,max}}$. (a) Normalized current noise PSD obtained with $V_{\text{read}} = -0.1$ V in the HRS upon increasing $V_{\text{reset,max}}$ from 2.0 V to 2.5 V in 0.1-V increments. Predominantly, $1/f^\alpha$ -type noise is observed, with $\alpha \approx 1.2$ (black line). (b) The frequency exponent α , (c) the normalized current noise PSD at 1 Hz, and (d) the HRS resistance as a function of $V_{\text{reset,max}}$. α is independent of $V_{\text{reset,max}}$, while the normalized PSD and the HRS resistance both increase as a function of $V_{\text{reset,max}}$. (e) The I - V characteristics of several intermediate resistive states (IRSs) obtained by incrementing $V_{\text{reset,max}}$. (f) A schematic of the CF evolution in Y_2O_3 during the successive incremental reset process. The CF cross section decreases with the increase of $V_{\text{reset,max}}$, thus increasing the DUT resistance as well as the normalized current noise PSD.

from the LRS, the CF is gradually reoxidized with increasing voltages, resulting in different IRSs with increasing resistances. At higher values of $V_{\text{reset,max}}$, we find that the device resistance is above that of the inverse conductance quantum, i.e., the resistance of a single atomic point contact, $G_0^{-1} = 12.9 \text{ k}\Omega$, which indicates that a spatial gap is induced in the filament. Upon further increase of the voltage, the gap widens and thereby deeper resets, resulting in higher resistance, can be achieved. In HfO_{2-x} and TaO_x -based RRAM, both the noise magnitude and the frequency exponent show a pronounced resistance dependence. In the work of Yu *et al.* [27], the authors report on the fluctuation spectroscopy analysis of HfO_{2-x} , where the change in the frequency exponent is attributed to the difference in the number of traps in the LRS and the HRS. It is explained that in the LRS, a high defect (trap) density in the CF leads to a $1/f$ -type frequency dependence, whereas the low number of defects in the ruptured part of the CF will lead to the appearance of isolated defects that act as traps and result in a $1/f^2$ or Lorentzian-type behavior. Similarly, the investigations of Yi *et al.* [26] on TaO_x describe $1/f$ -type spectra for metallic states with $R < 12.9 \text{ k}\Omega$, i.e., when the device is in the LRS, and they observe a $1/f^2$ frequency dependence for $R \geq 12.9 \text{ k}\Omega$ in the HRS. Contrary to these systems, we find a uniform noise behavior for Y_2O_3 -based RRAM. The frequency exponent α remains unchanged, whereas the noise magnitude and the resistance increase by incrementing $V_{\text{reset,max}}$, as shown in Figs. 2(b)–2(d). Therefore, the intrinsic high defect density of yttria serves as an explanation for and is consistent with the constant value of $\alpha \approx 1.2$ when compared to hafnium- or tantalum-oxide-based RRAM. A similar universality of α has been found in a Ag_2S -based CB-RAM device [30] operated in the ballistic and diffusive-transport regimes, i.e., without breaking the CF. There, the observed behavior is explained by internal structural fluctuations of Ag atoms between metastable atomic configurations within the nanofilament. In our case, however, the constant value of α at several IRSs with a wide range of resistances both below and above G_0 indicates that the physical mechanism responsible for the observed noise does not depend on the inner structure of the filament but, rather, originates from the CF environment, i.e., from processes in the insulating matrix surrounding the filament. The scaling of the noise magnitude with $V_{\text{reset,max}}$ and with the DUT resistance has been observed in other VCM systems and has been ascribed to the increased number of defects in the ruptured part of the CF [27,42]. Also, for most devices, it has been reported that the noise magnitude in the LRS is at least one order of magnitude smaller than in the HRS [32,43]. This is in good agreement with our findings that the noise magnitude scales with the resistance of the IRSs accessed by different values of $V_{\text{reset,max}}$. The observation of such universal $1/f$ -type noise suggests a large number of fluctuators with a wide distribution of capture and/or emission

times and can be correlated with the intrinsically high oxygen-defect density of yttrium oxide [20–22]. As previously demonstrated [17], the tunability of the resistance through reset voltage-amplitude control makes yttria-based RRAM a good candidate for neuromorphic and multibit applications. The evaluation of noise at these intermediate resistance levels is crucial, as electrical noise in read conditions can affect the IRS stability. Therefore, our current findings of low device-to-device variation, as well as the universal and reproducible resistance dependence of the noise, further solidify the place of yttria-based RRAMs amongst candidates for the implementation of artificial synapses and multilevel data storage.

Next, the cycle-to-cycle variation of the noise is investigated. Here, dc cycling is carried out with sweeping of the voltage to -2.5 V for set and $+2.5 \text{ V}$ for reset. Noise spectra are measured under read conditions ($V_{\text{read}} = -0.1 \text{ V}$) in the HRS after selected cycles, as shown in Fig. 3(a). In this case, the obtained spectra also show predominantly a $1/f^\alpha$ frequency dependence, with $\alpha \approx 1.2$. This finding is further confirmed by the endurance plots in Figs. 3(b)–3(d), where the frequency exponent α (shown in Fig. 3(b)), the normalized current noise PSD at 1 Hz (see Fig. 3(c)), and the HRS resistance (see Fig. 3(d)) are plotted as a function of the dc cycle number. Here, α again remains close to 1.2 during the 300 dc cycles, which can be attributed to the intrinsic abundance of oxygen vacancies specific to the sesquioxide structure of Y_2O_3 . Strikingly, we find that the current noise magnitude systematically decreases with dc cycling. A possible approach to explaining this behavior might be to correlate the noise magnitude with the device resistance, which gradually decreases during the first 150 dc cycles but then becomes stabilized, as shown in Fig. 3(d). This phenomenon is most likely due to the limited voltage values utilized in the reset process, as higher voltages, with which deeper reset can be achieved, are avoided to prevent the hard breakdown of the oxide layer. The noise magnitude, however, apparently continues to decrease even from cycle 150 to 300, also shown by the linear fit of the data in Fig. 3(c) (red line). Both the stabilization of resistance and the decrease of the noise magnitude can be explained by a so-called “training effect,” which we relate to the structural evolution and stabilization of the CF, as schematically shown in Fig. 3(e). According to this simplified model, at low cycle numbers, this is manifested as decreasing the rupture length. It is also known that, given the atomic size of the filament, even small atomic reconfigurations can result in relatively large resistance changes [29,44,45]. At this stage, with the shrinking of the spatial gap also comes a decrease in the number of active fluctuators (traps) in the proximity of the filament, which contribute to the electronic noise, resulting in a decreasing current noise magnitude. As the number of dc cycles increases, the filament stabilizes, thus, the reset process will induce only an incomplete rupture,

FIG. 3. The effect of dc cycling on the low-frequency noise behavior. (a) The normalized current noise spectra measured at $V_{\text{read}} = -0.1 \text{ V}$ after selected cycles; (b)–(d) The dc cycling dependence of the frequency exponent α , the normalized noise PSD at 1 Hz, and the HRS resistance, respectively. The frequency exponent does not exhibit any cycle dependence within the experimental error, whereas the noise PSD and HRS resistance both decrease over cycling. (e) A schematic of the CF evolution under dc cycling. The CF structure changes along the CF growth axis at low number of dc cycles. At a high cycle number, the CF is more stable with a larger cross section and switching occurs as a lateral change.

and resistive switching will happen via a cross-sectional change (the CF becomes narrower during reset and wider during set; see Fig. 3(e)). This behavior is also indicated by the HRS-resistance stabilization around G_0 in Fig. 3(d). Subsequent dc cycling in this stabilized state leads to driving the oxygen vacancies toward the filament, where they

are eventually consumed by it, increasing its lateral size. This process does not induce significant changes in the DUT resistance; however, the number and impact of potential fluctuators, i.e., oxygen vacancies acting as traps in the close vicinity of the CF, becomes smaller, resulting in the observed continued decrease of the noise magnitude. At the same time, due to the intrinsically high defect density of yttria, it is the overall effect of the high number of active fluctuators that dominates and thus the frequency dependence remains unaltered. It is also important to point out that, as we have previously reported [17], yttria-based devices show good endurance over more than 800 dc switching cycles with a minimum resistance ratio of over 10^2 , much higher than the number of cycles necessary to achieve the beneficial effect of lowering the current noise magnitude. These results point toward the exciting possibility of reducing intra- and interdevice variability, as well as the improvement of noise immunity through dc training in filamentary RRAM devices.

IV. CONCLUSION

In summary, low-frequency noise measurements on Y₂O₃-based RRAM devices, an artificial-synapse-material candidate for neuromorphic applications, have revealed universal noise in terms of the distribution of spectral weight being independent of the device resistance, the applied reset voltage amplitude, and the number of dc switching cycles. We attribute this universality to the intrinsically high oxygen vacancy density in yttria, which is specific to its sesquioxide structure. The scaling of the normalized current noise with the device resistance upon increasing reset voltage amplitude indicates that the current fluctuations originate from charge-capture and -emission processes at trap sites in the vicinity of the conductive filament. The dc endurance training stabilizes the filament via the consumption of oxygen vacancies from its close vicinity, resulting in a reduced noise magnitude due to the decreasing number of active traps that affect filament conductivity. The observed low device-to-device variation under multistate operation conditions and the reduction of the noise magnitude via dc endurance training promote yttria-based RRAM for both nonvolatile memory and neuromorphic applications.

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