

Large $1/f$ noise of unipolar resistance switching and its percolating nature

S. B. Lee,¹ S. Park,² J. S. Lee,³ S. C. Chae,¹ S. H. Chang,¹ M. H. Jung,⁴ Y. Jo,²
B. Kahng,³ B. S. Kang,⁵ M.-J. Lee,⁶ and T. W. Noh^{1,a)}

¹ReCOE, Department of Physics and Astronomy, Seoul National University, Seoul 151-747, Republic of Korea

²Quantum Materials Research Team, Korea Basic Science Institute, Daejeon 305-333, Republic of Korea

³Department of Physics and Astronomy, Seoul National University, Seoul 151-747, Republic of Korea

⁴Department of Physics, Sogang University, Seoul 121-742, Republic of Korea

⁵Department of Applied Physics, Hanyang University, Ansan, Gyeonggi-do 426-791, Republic of Korea

⁶Samsung Advanced Institute of Technology, Suwon, Gyeonggi-do 440-600, Republic of Korea

(Received 11 August 2009; accepted 3 September 2009; published online 25 September 2009)

We investigated the $1/f$ noise of Pt/NiO/Pt capacitors that show unipolar resistance switching. When they were switched from the low to high resistance states, the power spectral density of the voltage fluctuation was increased by approximately five orders of magnitude. At 100 K, the relative resistance fluctuation S_R/R^2 in the low resistance state displayed a power law dependence on the resistance R : i.e., $S_R/R^2 \propto R^w$, where $w=1.6 \pm 0.2$. This behavior can be explained by percolation theory; however, at higher temperatures or near the switching voltage, S_R/R^2 becomes enhanced further. This large $1/f$ noise can be therefore an important problem in the development of resistance random access memory devices. © 2009 American Institute of Physics. [doi:10.1063/1.3237167]

Recently, unipolar resistance switching (RS) phenomena has attracted much attention, due to potential applications in resistance random access memory (RRAM).¹⁻⁴ From an application viewpoint, the signal-to-noise ratio of RRAM should be as large as possible. Additionally, measurements of noise can provide valuable information on the electronic transport and the microscopic switching mechanisms that cannot be acquired by other measurements.⁵⁻⁷ Despite its scientific and technological importance, there has been little investigation of the noise behavior of unipolar RS.

Here, we report on $1/f$ noise of unipolar RS by using Pt/NiO/Pt capacitors. When we applied a bias V to our samples, we observed that the power spectral density S_V of the low frequency voltage fluctuation was inversely proportional to frequency f . We found that the $1/f$ noise in the low resistance state (LRS) increased significantly as the corresponding resistance increased. Detailed behavior of the $1/f$ noise will be discussed within the framework of the percolation theory. We will also present some experimental data that cannot be understood by such classical theory.

We deposited polycrystalline NiO thin films on Pt-coated Si substrates using dc magnetron reactive sputtering. To measure the electrical properties and noise, we fabricated Pt/NiO/Pt capacitors by depositing Pt top electrodes with an area of $50 \times 50 \mu\text{m}^2$ onto the NiO layer. Figure 1(a) shows current-voltage (I - V) curves of a Pt/NiO/Pt capacitor, which exhibits unipolar RS. The NiO capacitor was highly insulating in the pristine state (PS), marked by the black circles. Immediately after the forming process at V_F , it enters the LRS. When V was increased above V_R , it switched from the LRS to the high resistance state (HRS). This switching behavior was termed a reset process. As we increased V further, reaching V_S , the capacitor switched back into the LRS. This was termed a set process. To prevent permanent damage (i.e., complete dielectric breakdown) during both forming and set

processes, we used a compliance current I_{comp} of 1 mA.

We applied a bias to the Pt/NiO/Pt capacitor using a source-meter unit (Keithley 2601) and measured the noise intensity S_V using a spectrum analyzer (Agilent E4448A).⁸ To amplify the S_V signal, two kinds of low-noise differential amplifiers were used (NF SA-400F3 for LRS and ITHACA 1201 for HRS and PS). We also used a Wheatstone bridge, in which one of the resistors was replaced with our Pt/NiO/Pt capacitor, to measure the small noise signal with a high sensitivity. As shown in Fig. 1(c), we observed that the S_V signals in all of the resistance states had $1/f^\alpha$ dependence under an external bias. Since $\alpha \approx 1$, we will refer to the S_V signal as $1/f$ noise from now on.

Even when $V=0$ V, there should be f -independent voltage fluctuations S_V^{th} (i.e., Johnson noise^{5,7}). According to the fluctuation-dissipation theorem, the corresponding S_V^{th} should be proportional to the resistance R and temperature T . The theoretical values of S_V^{th} for the PS and HRS are shown by the dashed lines in Fig. 1(c). Our experimental S_V^{th} for the PS and HRS are in good agreement with the theoretical predictions. The theoretical S_V^{th} for the LRS should be smaller than that for the HRS by five orders of magnitude; however, this was below the experimental sensitivity. We also investigated the V -dependence of S_V in the LRS. In the Ohmic conduction region for most metals, S_V should follow Hooge's equation (i.e., $S_V \propto V^2$).⁵⁻⁷ As shown by the blue line in Fig. 1(b), $S_V \propto V^2$ in the LRS with $V < 0.15$ V, where Ohmic conduction occurs. These confirmations indicate that our $1/f$ noise measurements were highly reliable.

Figure 1(b) shows the S_V -values of a Pt/NiO/Pt capacitor, measured at room temperature with $f=100$ Hz. For any resistance state, as V increases, S_V increases slowly when V is small, but experiences a rapid increase near the corresponding switching voltage. We can readily convert S_V to the power spectral density of the resistance fluctuation S_R using $S_R = S_V/I^2$ (Ref. 7). While R^{HRS} was larger than R^{LRS} by five

^{a)}Electronic mail: twnoh@snu.ac.kr.

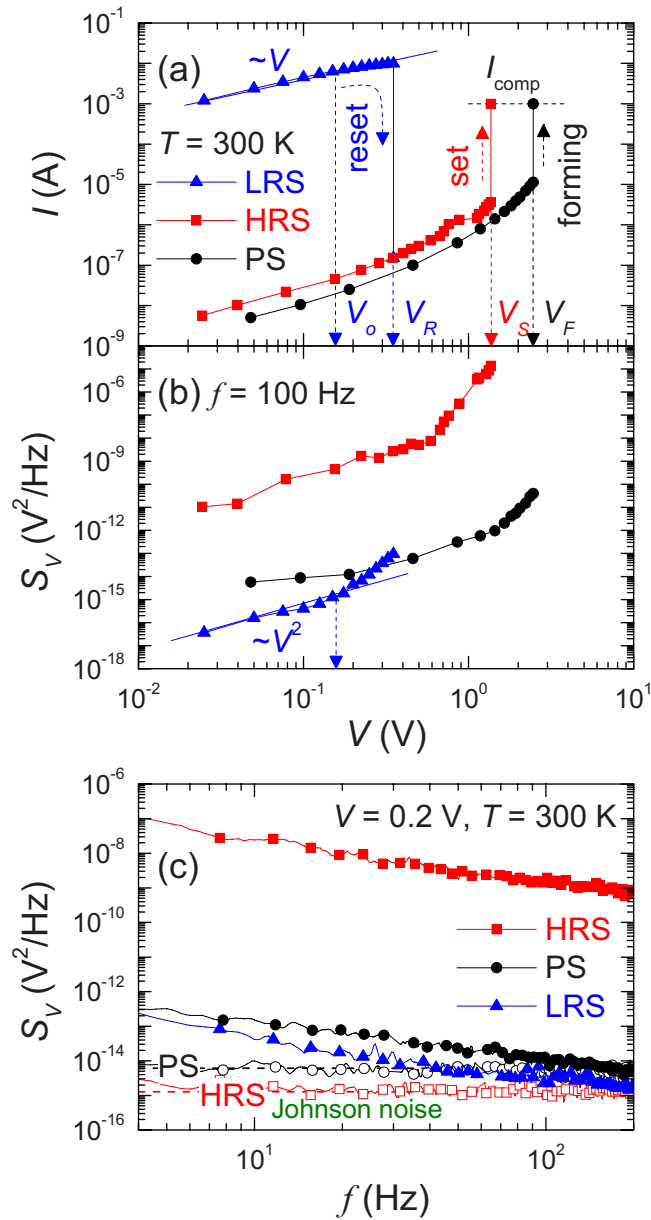


FIG. 1. (Color online) (a) I - V characteristics of a Pt/NiO/Pt capacitor. The LRS shows Ohmic behavior ($I \propto V$, blue solid line) below $V_o = 0.15$ V. (b) V -dependence of S_V . At the Ohmic region, S_V^{LRS} follows Hooge's equation, i.e., $S_V \propto V^2$. As V approaches V_R , V_S , and V_F , respectively, S_V starts to increase rapidly. (c) f -dependence of S_V . With $V = 0$ V, S_V^{th} of each resistance state (open symbols) follows theoretical values of Johnson noise (the dashed lines). With $V > 0$ V, each state shows $1/f^\alpha$ noise (solid symbols) with α close to unity. Note that $S_V^{\text{PS}} > S_V^{\text{HRS}} > S_V^{\text{LRS}}$ under $V = 0$ V and $S_V^{\text{HRS}} > S_V^{\text{PS}} > S_V^{\text{LRS}}$ under $V > 0$ V.

orders of magnitude, S_R^{HRS} was found to be larger than S_R^{LRS} by 15 orders of magnitude at 0.2 V.

Why does the $1/f$ noise become so large in the HRS? It is widely accepted that unipolar RS can occur due to the formation and rupture of conducting filaments under an applied electric field.^{1-4,9-13} The LRS is characterized by the conducting filaments making percolating channels across the sample. When the channels become broken by Joule heating, the HRS applies. $1/f$ noises in percolating material systems have been treated by classical percolation theories.^{7,8,14} It is known that S_R probes the fourth moments of current distributions in the percolating network.^{7,14} More explicitly, S_R/R^2 can be written as

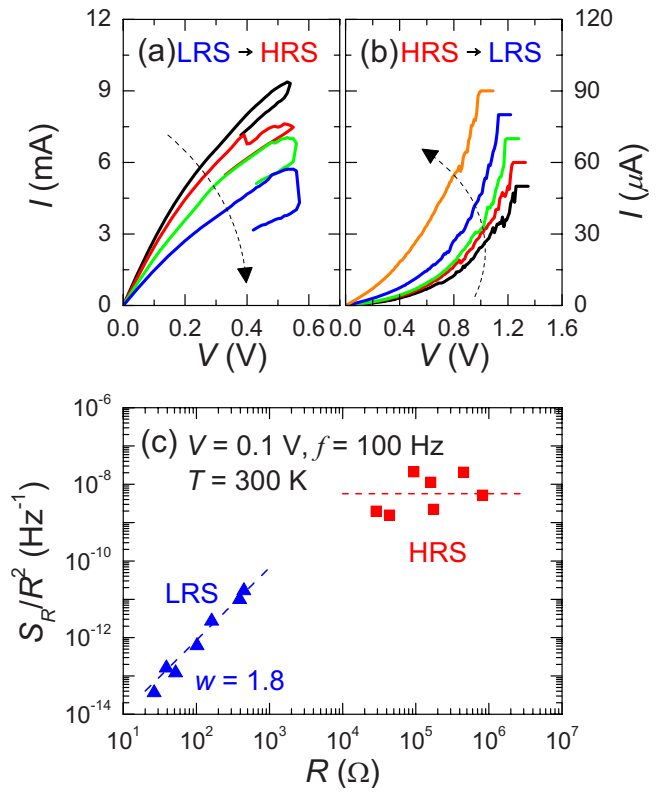


FIG. 2. (Color online) Obtaining multilevels of (a) the LRS and (b) the HRS by carefully controlling V . (c) The R -dependence of S_R/R^2 in the LRS and HRS. While S_R/R^2 -values of the LRS are proportional to R^w with $w = 1.8 \pm 0.3$, the HRS shows very large S_R/R^2 , scattered around $10^{-8} Hz^{-1}$.

$$\frac{S_R}{R^2} = \frac{\sum \rho_b^{2,4}}{(\sum r_b i_b^2)^2}, \quad (1)$$

where i_b is the current flowing through each filament b , whose resistance value is $r_b + \delta r_b$, and $\rho_b^2 = \langle \delta r_b \delta r_b \rangle$ is determined by the mechanism of fluctuations in the filament, which typically varies as the $1/f$ type. As the fraction, p , of conducting filaments approaches to the percolation threshold, p_c , S_R should diverge as $S_R \propto (p - p_c)^{-\kappa}$, where κ is the noise exponent.^{7,8} This divergence near p_c might explain the drastic increase in S_R in the HRS.

We performed quantitative studies on the relative noise S_R/R^2 . According to percolation theory,^{7,8,14}

$$\frac{S_R}{R^2} \propto R^w \quad \text{at } p > p_c. \quad (2)$$

To measure S_R for numerous resistance values in the LRS near p_c , we generated some multilevel LRS by carefully increasing V near V_R [Fig. 2(a)].¹⁵ Similarly, several multilevel HRS were also generated [Fig. 2(b)]. At each multilevel state, we measured S_V at room temperature with $V = 0.1$ V and plotted the corresponding S_R/R^2 -value in Fig. 2(c). In the LRS, S_R/R^2 has the power law dependence with $w = 1.8 \pm 0.3$. In the HRS, S_R/R^2 becomes very large and scattered around $10^{-8} Hz^{-1}$.

To obtain further insight, we measured S_R in the LRS by varying T and V . Each point in Fig. 3 was obtained at different LRS. As shown in Fig. 3(a), with a decrease in T from 300 to 100 K and with $V = 0.1$ V, w decreased from 1.8 to 1.6. However, as shown in Fig. 3(c), with $V = 0.3$ V, w de-

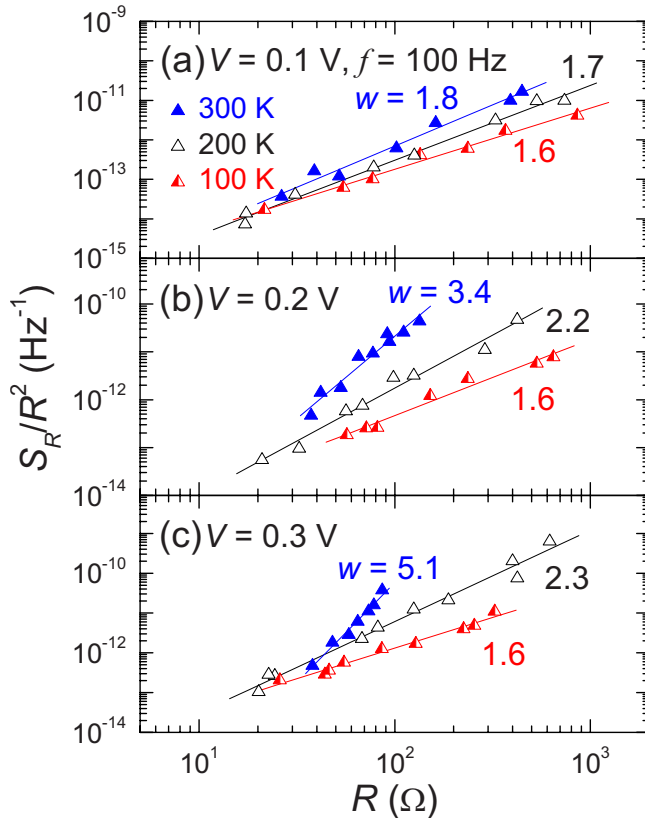


FIG. 3. (Color online) The R -dependence of S_R/R^2 in the LRS for $V=(a)$ 0.1, (b) 0.2, and (c) 0.3 V. The red half-filled, black open, and blue solid triangles show S_R/R^2 at 100, 200, and 300 K, respectively. Note that w increases with increasing V and T .

creased drastically from 5.1 to 1.6. It should be noted that all of the values of w at 100 K were nearly the same, i.e., $w=1.6 \pm 0.2$, regardless of V .

Note that percolation theory deals with the enhancement of S_R/R^2 due to the current distribution in the percolating network.^{7,8,14} Specifically, it deals with a purely geometrical effect, assuming that the resistance fluctuation in the conducting filament networks should be the same. So, the observation that $w=1.6 \pm 0.2$ at low T may be related to a geometric effect. According to classical lattice percolation theory,^{7,8} $0.82 < w < 1.05$. For two-dimensional (2D) semicontinuous metal films,^{7,8} it has been reported that $1.2 < w < 2.0$. To explain the unipolar RS phenomena, we recently developed a new type of percolation model, termed “the random circuit breaker (RCB) network model,” where fuse and antifuse actions occur depending on the voltage applied to a filament.¹¹ We performed 2D numerical simulations using the RCB network model and found $w=1.5 \pm 0.3$. This value agrees quite well with our observed w at 100 K, suggesting that the low temperature S_R/R^2 should be related to the current distribution in the percolating network.

The drastic increase in S_R/R^2 in the LRS near the reset voltage V_R (especially at high T) should be due to additional effects. The first possibility may be Joule heating effects, especially near the weakest bond, where most of the current will flow.^{1,3,4,9,13,15} To investigate this, we included Joule heating and associated thermal dissipation processes in the

RCB network model.^{11,13,16} However, we still obtained $w=1.5 \pm 0.3$. Another possibility is motion of ions, such as oxygen, near the RS, which are currently discussed by many workers.^{1,3,4} At high T and near V_R , such ionic motion could become important and work as an extra source for $1/f$ noise.⁵⁻⁷ Further systematic investigations are highly desirable to investigate this phenomenon.

A further important point related to the noises in the unipolar RS is that at $V=V_S$, $S_V^{HRS} \approx 10^{-4}$ V²/Hz [Fig. 1(b)]. This is much larger than the S_V -value of phase-change random access memory¹⁷ (where $S_V \approx 10^{-10}$ V²/Hz), and that of magnetic random access memory¹⁸ (where $S_V \approx 10^{-15}$ V²/Hz). For RRAM applications, the large noise issue should be carefully addressed.

In summary, we performed systematic investigations into the $1/f$ noise of unipolar RS. Our experimental $1/f$ noise data at 100 K and low applied bias confirms our view that unipolar RS should occur by the percolation process of conducting filaments. However, at higher temperature or near the switching voltages, $1/f$ noise became further enhanced, indicating the existence of additional noise sources. Our work indicates that $1/f$ noise of unipolar RS is an important issue and should be investigated further for RRAM device applications.

This research was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (Grant No. 2009-0080567). B.K. and J.S.L. were supported by the KOSEF grant funded by the MOST (Grant No. R17-2007-073-01001-0).

¹R. Waser and M. Aono, *Nature Mater.* **6**, 833 (2007).

²G. I. Meijer, *Science* **319**, 1625 (2008).

³M.-J. Lee, S. Han, S. H. Jeon, B. H. Park, B. S. Kang, S.-E. Ahn, K. H. Kim, C. B. Lee, C. J. Kim, I.-K. Yoo, D. H. Seo, X.-S. Li, J.-B. Park, J.-H. Lee, and Y. Park, *Nano Lett.* **9**, 1476 (2009).

⁴R. Waser, R. Dittmann, G. Staikov, and K. Szot, *Adv. Mater. (Weinheim, Ger.)* **21**, 2632 (2009).

⁵P. Dutta and P. M. Horn, *Rev. Mod. Phys.* **53**, 497 (1981).

⁶M. B. Weissman, *Rev. Mod. Phys.* **60**, 537 (1988).

⁷Sh. Kogan, *Electronic Noise and Fluctuations in Solids* (Cambridge University Press, Cambridge, 1996).

⁸Y. Song, S.-I. Lee, and J. R. Gaines, *Phys. Rev. B* **46**, 14 (1992).

⁹B. J. Choi, S. Choi, K. M. Kim, Y. C. Shin, C. S. Hwang, S.-Y. Hwang, S.-S. Cho, S. Park, and S.-K. Hong, *Appl. Phys. Lett.* **89**, 012906 (2006).

¹⁰S. B. Lee, S. C. Chae, S. H. Chang, C. Liu, C. U. Jung, and D.-W. Kim, *J. Korean Phys. Soc.* **51**, S96 (2007).

¹¹S. C. Chae, J. S. Lee, S. Kim, S. B. Lee, S. H. Chang, C. Liu, B. Kahng, H. Shin, D.-W. Kim, C. U. Jung, S. Seo, M.-J. Lee, and T. W. Noh, *Adv. Mater. (Weinheim, Ger.)* **20**, 1154 (2008).

¹²S. B. Lee, S. C. Chae, S. H. Chang, J. S. Lee, S. Seo, B. Kahng, and T. W. Noh, *Appl. Phys. Lett.* **93**, 212105 (2008).

¹³S. H. Chang, J. S. Lee, S. C. Chae, S. B. Lee, C. Liu, B. Kahng, D.-W. Kim, and T. W. Noh, *Phys. Rev. Lett.* **102**, 026801 (2009).

¹⁴R. Rammal and A.-M. S. Tremblay, *Phys. Rev. Lett.* **58**, 415 (1987).

¹⁵S. B. Lee, S. C. Chae, S. H. Chang, J. S. Lee, S. Park, Y. Jo, S. Seo, B. Kahng, and T. W. Noh, *Appl. Phys. Lett.* **93**, 252102 (2008).

¹⁶D. Lubin, I. Goldhirsch, and Y. Gefen, *Phys. Rev. B* **38**, 5899 (1988).

¹⁷P. Fantini, A. Pirovano, D. Ventrice, and A. Redaelli, *Appl. Phys. Lett.* **88**, 263506 (2006).

¹⁸W. K. Park, J. S. Moodera, J. Taylor, M. Tondra, J. M. Daughton, A. Thomas, and H. Brückl, *J. Appl. Phys.* **93**, 7020 (2003).