ADVANCED MATERIALS

DOI: 10.1002/adma.200702024

Random Circuit Breaker Network Model for Unipolar Resistance Switching**

By *Seung Chul Chae, Jae Sung Lee, Sejin Kim, Shin Buhm Lee, Seo Hyoung Chang, Chunli Liu, Byungnam Kahng, Hyunjung Shin, Dong-Wook Kim, Chang Uk Jung, Sunae Seo, Myoung-Jae Lee,* and *Tae Won Noh**

The existence of reversible resistance switching (RS) behaviors induced by electric stimulus has been known for some time,[1–3] and these intriguing physical phenomena have been observed in numerous materials, including oxides.[4–17] As conventional charge-based random access memory is expected to face a size limit in the near future, a surge of renewed interest has been developed in RS phenomena for possible applications in small nonvolatile memory devices called resistance random access memory (RRAM).

Of particular interest is unipolar RS, which shows the RS at two values of applied voltage of the same polarity.[15–17] The unipolar RS exhibits a much larger resistance change than other RS phenomena, and this greatly simplifies the process of reading the memory state. When fabricated with oxide *p-n* diodes, memory cells using unipolar RS can be stacked vertically, which has the potential for dramatically increasing memory density.[18] Therefore, unipolar RRAM may be a good candidate for multi-stacked, high density, nonvolatile memory.

The most important scientific and technical issues concerning unipolar RS are how it works and the identification of its controlling parameters. Some studies have reported that unipolar RS comes from a homogeneous/inhomogeneous transition of current distribution,[19] while others maintain that it comes from the formation and rupture of conducting filaments.[13,20] Even with recent extensive studies on unipolar RS, its basic origin is still far from being understood. In addition, no model exists that actually explains how the reversible switching can occur at two values of applied voltage. This lack of a quantitative model poses a major barrier for unipolar RRAM applications.

In this study, we describe RS behavior in polycrystalline TiO_2 film. To explain the basic mechanism of unipolar RS behavior, we propose a new percolation model based on a network of "circuit breakers" with two switchable metastable states. The random circuit breaker (RCB) network model can explain the long-standing material issue of how unipolar RS occurs. This simple percolation model is different from the conventional percolation models, which have dealt only with static or irreversible dynamic processes. In addition, the RCB network model provides an indication of how to overcome the substantial distribution of switching voltages, which is currently considered the most serious obstacle to practical unipolar RRAM applications.[21]

The unipolar RS phenomenon can be explained by the current (I)-voltage (V) curves in Figure 1a, which are derived from measurements of our polycrystalline TiO_2 thin capacitors. At the pristine state (green dot), they are in an insulating state. As the external voltage V_{ext} increases from zero and reaches a threshold voltage $V_{forming}$, a sudden increase occurs in the current. If the current is not limited to a certain value, here called the compliance current I_{comp}, the TiO_2 capacitor would experience a dielectric breakdown and be destroyed. However,

[*] Prof. T. W. Noh, S. C. Chae, S. B. Lee, S. H Chang, Dr. C. Liu
ReCOE & FPRD, Department of Physics and Astronomy
Seoul National University
Seoul 151-747 (Korea)
E-mail: twnoh@snu.ac.kr

J. S. Lee, Prof. B. Kahng
Department of Physics and Astronomy
Seoul National University
Seoul 151-747 (Korea)

S. Kim, Prof. H. Shin
Center for Materials and Processes of Self-Assembly
School of Advanced Materials Engineering
Kookmin University
Seoul 136-702 (Korea)

Prof. D.-W. Kim
Department of Applied Physics
Hanyang University
Ansan, Gyeonggi-do 426-791 (Korea)

Prof. C. U. Jung
Department of Physics
Hankuk University of Foreign Studies
Yongin, Gyeonggi-do 449-791 (Korea)

Dr. S. Seo,[+] Dr. M.-J. Lee[+]
Samsung Advanced Institute of Technology
Suwon 440-600 (Korea)

[+] Present address: Division of Nano Sciences and Department of Physics, Ewha Womans University, Seoul 120-570 (Korea).

[**] S. C. C. and J. S. L. contributed equally to this work. This study was supported principally by a Creative Research Initiative (Functionally Integrated Oxide Heterostructures) from the Korean Ministry of Science and Technology (MOST) and the Korea Science and Engineering Foundation (KOSEF), and in part by Samsung Electronics. B. K. and J. S. L were supported by the KOSEF grant funded by the MOST (No.R17-2007-073-01001-0). H. S. and S. K. would like to thank the Center for Materials and Processes of Self-assembly (R11-2005-048-00000-0) and National Research Lab (R0A-2007-000-20105-0) Program in the MOST/KOSEF ERC program. Supporting Information is available from Wiley InterScience or from the author.

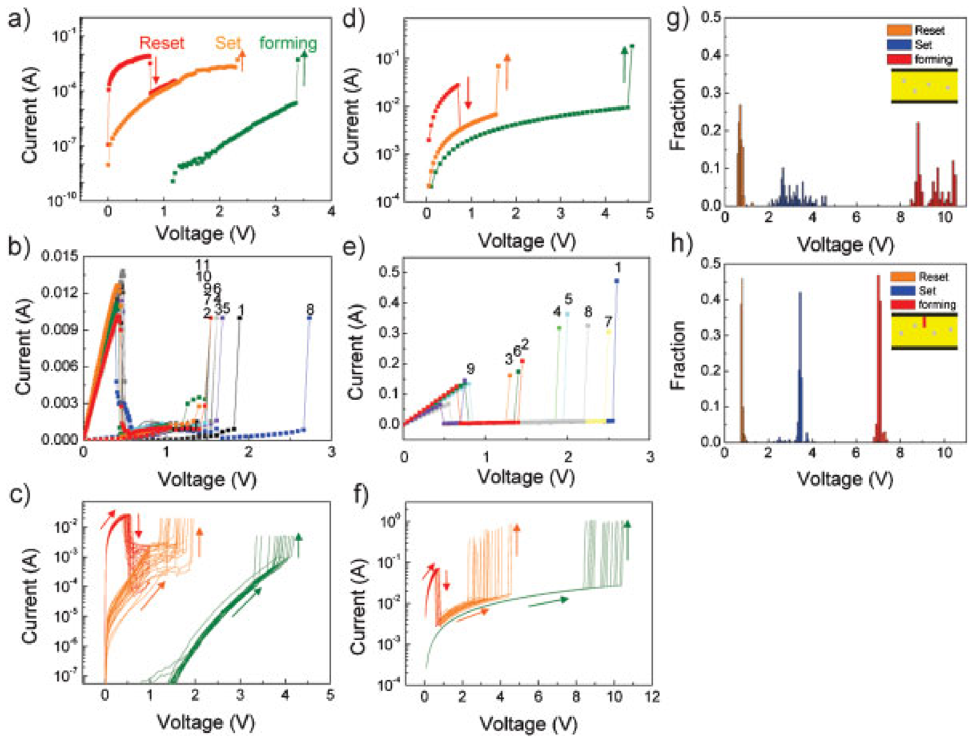

Figure 1. *I–V* measurements and simulation results of unipolar resistance switching (RS) behaviors. a) Unipolar RS under the same bias polarity but with different magnitude, in polycrystalline TiO_2 thin film. b) Distributions of V_{RESET} and V_{SET} for a TiO_2 cell with the same I_{comp} value of 0.01 A. c) Wide distributions of $V_{forming}$, V_{RESET}, and V_{SET} of the TiO_2 cells fabricated under identical conditions. d) The simulation result for the unipolar RS using the RCB network model. e) The simulated result of a wide distribution of V_{RESET} and V_{SET} in a particular pristine configuration. f) The simulated result of wide distributions of $V_{forming}$, V_{RESET}, and V_{SET} for numerous pristine configurations with $p_{init} = 0.5\%$. g) Wide distributions in $V_{forming}$, V_{RESET}, and V_{SET} without any defective circuit breakers. h) The reduction in all distributions of $V_{forming}$, V_{RESET}, and V_{SET} with four aligned defective circuit breakers introduced at one electrode.

when I_{comp} is set properly, the TiO_2 capacitor goes into the low resistance state (LRS) in what is often called the "forming operation".[15] In the LRS (red dots), the current is rather high for any given voltage. As V_{ext} increases again from zero, a sudden drop occurs in the current, Reset operation, at the Reset voltage V_{RESET}. This high resistance state (HRS) is metastable, and the device remains in the same state for $0 \leq V_{ext} \leq V_{SET}$. In the HRS (orange dots), the current is rather low. As V_{ext} increases from zero once more, a sudden increase occurs in the current, Set operation, at the Set voltage V_{SET}. In this case, we also need to limit I_{comp} to avoid a dielectric breakdown. The resulting LRS is also metastable and the device remains in this state for $0 \leq V_{ext} \leq V_{RESET}$. These LRS and HRS can be used as the binary states for memory applications.

Figure 1b shows a series of *I–V* curves from repeated Set and Reset operations for a single TiO_2 capacitor. The I_{comp} value for Set was fixed at 10 mA, and the numbers on top of the Set curves indicate the repetition numbers. Although we used the same capacitor, distributions occur in V_{RESET} and V_{SET}. Figure 1c shows a series of *I–V* curves for numerous TiO_2 capacitors prepared under the same fabrication conditions. Wide distributions in $V_{forming}$, V_{RESET}, and V_{SET} also occur, which makes it difficult to fabricate reliable RRAM devices. Indeed, the wide distributions in V_{RESET} and V_{SET} are considered the major obstacles to practical RRAM applications.[21]

In spite of these wide distributions of V_{RESET} and V_{SET}, all the *I–V* curves in Figures 1b and 1c show an intriguing phenomenon. Note that the resistance changes at V_{RESET} and V_{SET} are nearly the same at least in terms of being the same order of magnitude. We will call this phenomenon the universal resistance change. Considering the rather wide distribution of V_{RESET} and V_{SET} values, the existence of this universal resistance is rather surprising.

We performed conductive atomic force microscopy (C-AFM) studies shown schematically in Figure 2a to gain further insight into unipolar RS. The C-AFM images as well as the *I–V* characteristics provide local information about the current flow paths through the TiO_2 film with high lateral resolution. Figure 2b shows the local *I–V* curves taken at one point on the surface. By applying the external voltage to the C-AFM tip as a top electrode, the unipolar RS operations of forming, Reset, and Set could be triggered.

The C-AFM images provide evidence of the formation and rupture of conducting channels during RS in our TiO_2 film. Note that for all the current maps in this study, we applied a bias of 0.1 V to the C-AFM tip. First, to obtain the LRS after the

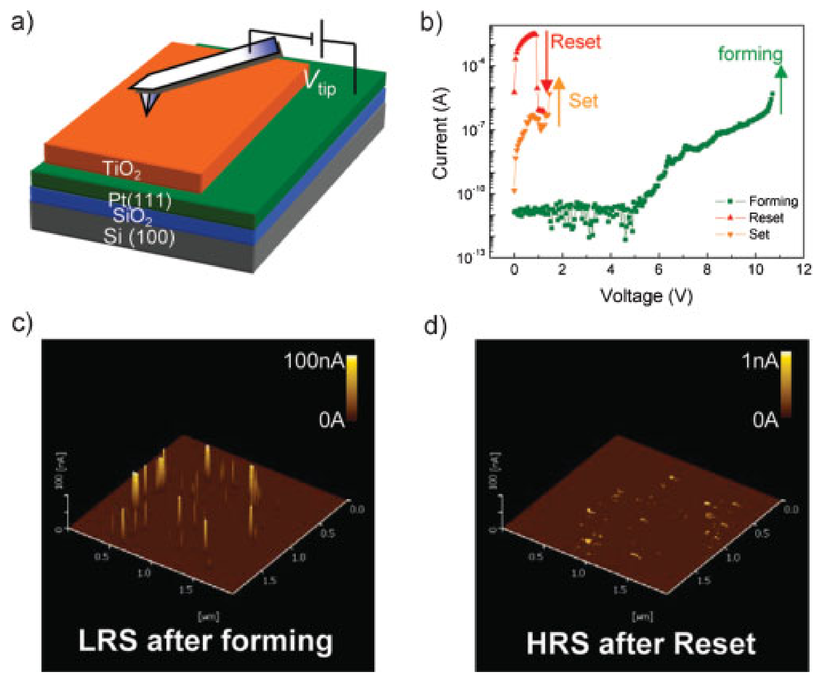

Figure 2. AFM measurements (C-AFM) on TiO_2 films. a) Schematic diagram of the C-AFM measurements. b) The *I*–*V* curves using the conducting AFM tip as a top electrode, clearly show the forming, Reset, and Set operations. c) Mapping of the current flow through the surface just after the forming operation with $V_{tip} = 8$ V shows locally distributed conducting regions. d) After the Reset operation with $V_{tip} = 1$ V, the TiO_2 surface in the HRS state shows that the locally distributed conducting regions disappear.

forming operation, we scanned the film with a bias of 8.0 V, measured the current flowing, and made a current map. As shown in Figure 2c, the C-AFM current map of the electrically formed surface, that is, the LRS, reveals several highly conductive spots, typically with diameters of about 3–10 nm. Second, we obtained the HRS by applying the Reset voltage of 1.0 V. Figure 2d shows that all the conducting spots in the HRS had disappeared. Third, by repeating the Reset and Set operations, we found that some of the conductive spots in the LRS were variable.[22]

To explain unipolar RS, we propose a new percolation model, the RCB network model. Figure 3a is a schematic representation of a bond percolation network composed of circuit breakers. Each circuit breaker can have either one of two resistance values, r_h or r_l, where $r_h \gg r_l$. Let us assume that the circuit breakers that exhibit r_h are in the off-state, marked as the black symbols. Conversely, the breakers that have a resistance of r_l are assumed to be the on-state, and are marked in red. As shown in Figure 3b, we assume that the resistance state switches depending on the magnitude of voltage Δv applied across the circuit breaker.

$$\text{For the on-state circuit breaker, on-state} \rightarrow \text{off-state when } \Delta v > v_{off} \tag{1}$$

$$\text{For the off-state circuit breaker, off-state} \rightarrow \text{on-state when } \Delta v > v_{on}, \tag{2}$$

where $v_{on} \gg v_{off}$. Note that these actions are similar to turning a real circuit breaker off and on, and could also correspond to the rupture and formation of a small segment of conductive filament.

In the computer simulations, the on-state circuit breakers in the pristine state were chosen randomly with a number fraction of p_{init}. Then, we started to increase V_{ext} and perform simulations. If switching occurred in at least one circuit breaker, we reevaluated the Δv distribution, and checked the switching conditions again, i.e., (1) and (2) above. We found that a switching in one circuit breaker triggered an avalanche of switchings in other circuit breakers nearby. We repeated the iteration operations until reaching a static state for the Reset operation. However, for the Set and the forming operations, we stopped the iteration process when the current flowing through the network became larger than a specified value of I_{comp}.

Figure 1d shows RCB network simulation results for the *I*–*V* curves for two-dimensional (2-D) lattices of 50×20 breakers, with $r_h = 1000$ Ohm cm, $r_l = 1$ Ohm cm, and $p_{init} = 0.5\%$. Note that at the expense of much longer simulation times, a larger value of r_h/r_l and three-dimensional simulations could provide a larger difference between the HRS and LRS, which would be closer to experimental results. For most simulations, the value of I_{comp} was fixed at 0.5 A. These 2-D simulation results can explain the observed unipolar RS behaviors in Figure 1a. In addition, although not shown here, this model can also explain other characteristics of the unipolar RS such as the *I*–*V* duali-

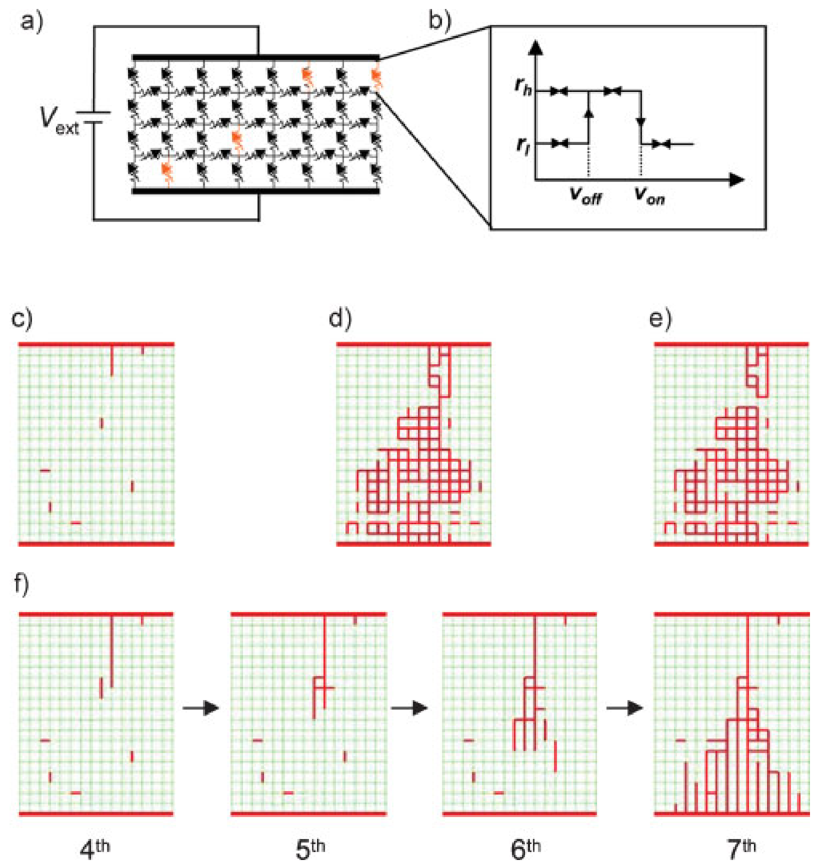

Figure 3. Detailed circuit breaker configurations in the RCB networks. a) Schematic diagram of the RCB network composed of circuit breakers. b) Detailed operations with switchable bistable states of circuit breakers. c) Pristine state with a few circuit breakers in the on-state. d) The LRS with a percolating cluster of circuit breakers in the on-state formed between the top and bottom electrodes. e) The HRS with a broken link; an off-state circuit breaker switched from the on-state. f) The configurations after the 4th, 5th, 6th, and 7th iterations during the forming operation show that the percolating cluster growth occurs as an avalanche process. These snapshots also indicate that the growth has a preferred direction, parallel to the applied electric field.

ty,[19] dependence of V_{SET} on I_{comp},[16] and observation of multilevel switching.[23] The success of the RCB model demonstrates that the basic operations of unipolar RS can be described as self-organized collective processes occurring in the network of circuit breakers with several simple rules for reversible switching.

Figure 1e displays a series of *I–V* characteristics, which are simulated by repeating Reset and Set operations for a particular pristine configuration. Figure 1f shows a series of *I–V* curves, which are simulated for numerous pristine configurations with $p_{init} = 0.5\%$. Note that the RCB network model can explain the wide distributions in $V_{forming}$, V_{RESET}, and V_{SET}. In addition, the amounts of resistance change at V_{RESET} and V_{SET} are nearly the same, supporting the observed universal resistance changes in Figure 1b and 1c.

We can obtain further insight by studying detailed configurations of RCB networks. In Figure 3c–f, we show only the region where a percolating path will be formed in the LRS. In the Supplementary section, we present the detailed simulation method and video clips for time-dependent circuit breakers configuration changes for the whole network, and the associated *I-V* curves for the forming and Reset/Set operations. Figure 3c indicates that only a few on-state circuit breakers exist in the pristine state. As Figure 3d shows, when V_{ext} reaches $V_{forming}$, the on-state circuit breakers form a percolating path and the network enters the LRS. For most simulations with 50×20 lattices, we found that only one percolating path was formed, and that the path was long in the vertical direction. This localized nature of the percolating path explains why the observed RS properties of the unipolar RRAM are weakly sensitive to the electrode size.[19,24] Note that size-insensitive properties of RS indicate the potential for RRAM to overcome the present charge-based memory scaling limit.[4] As shown in Figure 3e, when V_{ext} starts to increase from zero and reaches V_{RESET}, the percolating path breaks and the network enters the HRS.

A comparison of Figure 3d and 3e shows that the HRS can be formed by turning off a few circuit breakers in the percolating path. The universal resistance changes presented in Figure 1b and 1c can be explained by the detailed structure of the percolating cluster near the percolation threshold. Consider the simple case, where r_h is infinite and the size of network is also infinite. As shown in Figure 3d, the percolating cluster is composed of singly connected bonds or links, and multiply connected bonds or "blobs." Near the percolation threshold, a bond called the "hottest bond" exists through which all the current flows.[25] If we cut the hottest bond, the network will not allow the current to flow, which corresponds to the HRS in Figure 3e. Therefore, the connectivity of the hottest bond results in the universal resistance change, which is a hallmark of the percolation process.[26]

Note that the formation of percolating clusters during the forming and the Set operations should occur in self-organized avalanche processes. Figure 3f contains snapshots of the simulation after the 4th, 5th, 6th, and 7th iterations, which indicate the formation of a percolating cluster at $V_{ext} = V_{forming}$. During the first three iterations, only a small change was evident. However, after the 5th iteration, the largest cluster started to grow abruptly. This avalanche process resulted in a percolating cluster, which was long in the vertical direction. Note that in the Set operation, the percolating cluster will be created in a similar fashion, but starting from the HRS, which is to the left of the last Reset operation. Since more circuit breakers are already turned on, V_{SET} should be smaller than $V_{forming}$.

The RCB network model presents a new type of percolation problem, which can provide insights into reversible dynamic processes involving bistable states. While many percolation problems have been thoroughly investigated, most of them fall into two groups. Those with a cluster topology and structure are static in nature. Others are of a steady-state type, including diffusion of percolating clusters and electrical conductivity.[26] A few dynamic percolation problems have been studied, which include the breakdown of random media, also called the "random fuse model,"[25] and hydrodynamic dispersion in random media.[27] These models deal with irreversible dynamic processes, which always proceed in one direction in time. However, in our RCB network model, we are dealing with reversible dynamic processes involving bistable states. Our deterministic model can explain the seemingly chaotic behavior of V_{SET} in the repeated Reset and Set operations, shown in Figure 1b, with several simple reversible switching rules. In addition, our simulation results suggest that the detailed circuit breaker configuration in the HRS or LRS might depend on the initial condition in some deterministic way.

If nature follows the deterministic nature of our RCB network model, then this could provide ways to enhance RRAM performance. To demonstrate such possibilities, we will show one example, which introduces external defects at specific locations inside the RCB network. Figure 1g shows the probability distributions of $V_{forming}$, V_{RESET}, and V_{SET} for the simulations in Figure 1c, in which the on-state circuit breakers in the pristine state were chosen randomly. We repeated the same simulations with an additional artificial defect in the network by assuming that four aligned circuit breakers near the top electrode were always in the on-state, as shown in the inset of Figure 1h. Figure 1h shows that the distribution of $V_{forming}$ can be significantly improved.

In summary, we observed unipolar RS phenomena in polycrystalline TiO_2 capacitors. C-AFM studies demonstrated that local conducting channels could be formed, and that the resistance of a capacitor could be determined by the formation and rupture of conducting channels. We proposed the random circuit breaker network model to answer the long-standing material science question of how unipolar RS occurs. Unlike other percolation models, the RCB network model is unique since it can describe reversible dynamic processes involving two quasi-metastable states. In addition, it could provide new insights into the control of material parameters for fabricating high-performance unipolar RRAM devices.

Experimental

To fabricate the polycrystalline TiO_2 films used in this study, we deposited Ti metal films on $Pt/TiO_x/SiO_2/Si$ substrates by electron beam evaporation. During the Ti metal evaporation, we maintained the pressure inside our chamber at 3.0×10^{-7} Torr. Then we thermally oxidized the Ti metal films at 650 °C with a tube furnace. We maintained the oxygen flow rate at about 40 cc min^{-1}. We did not detect any Ti phase with X-ray diffraction studies using a synchrotron X-ray source, indicating that the film was completely TiO_2. For electrical measurements, we deposited Pt top electrodes using electron beam evaporation. We investigated RS behavior using a semiconductor parameter analyzer (4155c; Agilent Technologies, Palo Alto, CA, USA). We performed the C-AFM under a scanning probe microscope (SPA-400; SII Nanotechnology, Chiba, Japan). Using a cantilever coated with Pt as a conductive tip, we simultaneously and independently obtained topographical and electrical information from the insulating surfaces with a typical resolution of several nanometers.

For the computer simulation, we initially considered a square $M\times N$ lattice composed mostly of high-resistance bond r_h and partially of low-resistance bond r_l. We imposed the constraint that the $M\times N$ lattice be linked with a periodic boundary condition, and V_{ext} be applied along the vertical direction. With this initial lattice, we increased the V_{ext} from zero. At each voltage step, all the voltages across each bond Δv were calculated by solving the Laplace equation with the boundary condition for a fixed V_{ext}. In this simulation, we used the successive over-relaxation method to obtain the set of Δv.

Received: August 13, 2007
Revised: October 16, 2007

[1] G. Dearnaley, A. M. Stoneham, D. V. Morgan, *Rep. Prog. Phys.* **1970**, *33*, 1129.
[2] H. Biederman, *Vacuum* **1976**, *26*, 513.
[3] H. Pagnia, N. Sotnik, *Phys. Status Solidi A* **1989**, *111*, 387.
[4] K. Szot, W. Speier, G. Bihlmayer, R. Waser, *Nat. Mater.* **2006**, *5*, 312.
[5] A. Sawa, T. Fujii, M. Kawasaki, Y. Tokura, *Appl. Phys. Lett.* **2004**, *85*, 4073.
[6] S. Q. Liu, N. J. Wu, A. Ignatiev, *Appl. Phys. Lett.* **2000**, *76*, 2749.
[7] A. Beck, J. G. Bednorz, Ch. Gerber, C. Rossel, D. Widmer, *Appl. Phys. Lett.* **2000**, *77*, 139.
[8] M. J. Rozenberg, I. H. Inoue, M. J. Sanchez, *Phys. Rev. Lett.* **2004**, *92*, 178302-4.
[9] R. Dong, D. S. Lee, W. F. Xiang, S. J. Oh, D. J. Seong, S. H. Heo, H. J. Choi, M. J. Kwon, S. N. Seo, M. B. Pyun, M. Hasan, H. Hwang, *Appl. Phys. Lett.* **2007**, *90*, 042107-3.

[10] D. Lee, D.-J. Seong, I. Jo, F. Xiang, R. Dong, S. Oh, H. Hwang, *Appl. Phys. Lett.* **2007**, *90*, 122104-3.
[11] T. P. Chen, M. S. Tse, X. Zeng, *Appl. Phys. Lett.* **2001**, *78*, 492.
[12] M. Villafuerte, S. P. Heluani, G. Juárez, G. Simonelli, G. Braunstein, S. Duhalde, *Appl. Phys. Lett.* **2007**, *90*, 052105-3.
[13] W. R. Hiatt, T. W. Hickmott, *Appl. Phys. Lett.* **1965**, *6*, 106.
[14] K. C. Park, S. Basavaiah, *J. Non-Cryst. Solids* **1970**, *2*, 284.
[15] S. Seo, M. J. Lee, D. H. Seo, E. J. Jeoung, D.-S. Suh, Y. S. Joung, I. K. Yoo, I. R. Hwang, S. H. Kim, I. S. Byun, J.-S. Kim, J. S. Choi, B. H. Park, *Appl. Phys. Lett.* **2004**, *85*, 5655.
[16] C. Rohde, B. J. Choi, D. S. Jeong, S. Choi, J.-S. Zhao, C. S. Hwang, *Appl. Phys. Lett.* **2005**, *86*, 262907-3.
[17] H. Shima, F. Takano, Y. Tamai, H. Akinaga, I. H. Inoue, *Jpn. J. Appl. Phys.* **2007**, *46*, L57.
[18] M.-J. Lee, S. Seo, D.-C. Kim, S.-E. Ahn, D. H. Seo, I.-K. Yoo, I.-G. Baek, D.-S. Kim, I.-S. Byun, S.-H. Kim, I.-R. Hwang, J.-S. Kim, S.-H. Jeon, B. H. Park, *Adv. Mater.* **2007**, *19*, 73.
[19] I. H. Inoue, S. Yasuda, H. Akinaga, H. Takagi, ArXiv: cond-mat/0702564v1.
[20] J. F. Gibbons, W. E. Beadle, *Solid-State Electron.* **1964**, *7*, 785.
[21] D. C. Kim, M. J. Lee, S. E. Ahn, S. Seo, J. C. Park, I. K. Yoo, I. G. Baek, H. J. Kim, E. K. Yim, J. E. Lee, S. O. Park, H. S. Kim, U.-I. Chung, J. T. Moon, B. I. Ryu, *Appl. Phys. Lett.* **2006**, *88*, 232106-3.
[22] J.-B. Yun, S. Kim, S. Seo, M.-J. Lee, D.-C. Kim, S.-E. Ahn, Y. Park, J. Kim, H. Shin, *Phys. Status Solidi RRL* **2007**, *1*, 280.
[23] D. C. Kim, S. Seo, S. E. Han, D.-S. Suh, M. J. Lee, B.-H. Park, I. K. Yoo, I. G. Baek, H.-J. Kim, E. K. Yim, J. E. Lee, S. O. Park, H. S. Kim, U.-I. Chung, J. T. Moon, B. I. Ryu, *Appl. Phys. Lett.* **2006**, *88*, 202102-3.
[24] S. Seo, M. J. Lee, D. H. Seo, S. K. Choi, D.-S. Suh, Y. S. Joung, I. K. Yoo, I. S. Byun, I. R. Hwang, S. H. Kim, B. H. Park, *Appl. Phys. Lett.* **2005**, *86*, 093509-3.
[25] G. G. Batrouni, B. Kahng, S. Redner, *J. Phys. A: Math. Gen.* **1988**, *21*, L23.
[26] D. Stauffer, A. Aharony, in Introduction to Percolation Theory, Taylor & Francis, Washington, DC **1992**.
[27] A. H. Thompson, A. J. Katz, R. A. Raschke, *Phys. Rev. Lett.* **1987**, *58*, 29.