Magnetic field control of hysteretic switching in Co/Al_2O_3 multilayers by carrier injection

Alan Kalitsov, Ajeesh M. Sahadevan, S. Narayana Jammalamadaka, Gopinadhan Kalon, Charanjit S. Bhatia, Guangcheng Xiong, and Hyunsoo Yang

Citation: AIP Advances **1**, 042158 (2011); doi: 10.1063/1.3666854
View online: http://dx.doi.org/10.1063/1.3666854
View Table of Contents: http://aip.scitation.org/toc/adv/1/4
Published by the American Institute of Physics

Magnetic field control of hysteretic switching in Co/Al_2O_3 multilayers by carrier injection

Alan Kalitsov,[a] Ajeesh M. Sahadevan,[a] S. Narayana Jammalamadaka, Gopinadhan Kalon, Charanjit S. Bhatia, Guangcheng Xiong, and Hyunsoo Yang[b]

Department of Electrical and Computer Engineering, NUSNNI-Nanocore, National University of Singapore, 4 Engineering Drive 3, Singapore 117576, Singapore

(Received 21 October 2011; accepted 4 November 2011; published online 28 November 2011)

We propose a theoretical model of magnetic field dependence of hysteretic switching in magnetic granular system. The model is based on the self-trapped electrons mechanism. Our calculations show that the switching voltage may be significantly decreased with increasing the magnetic field. The underlying mechanism is the influence of the magnetic field on electron occupation of the conduction band, which depends on the materials used in magnetic granular system, concentration of magnetic granules in the insulating matrix, applied voltage, and the charge accumulation on the granules. We support our theoretical calculations by measuring the magnetic field dependence of resistive switching behaviour in Co/Al_2O_3 granular multilayers. Our experimental results are in qualitative agreement with the proposed theory. [doi:10.1063/1.3666854]

Resistive switching (RS) is a dramatic change in resistance of various metal-insulator systems induced by the electric field. Threshold RS is a switching between high resistance states (HRS) and low resistance states (LRS) when only one stable resistance state is preferable with no applied voltage. RS phenomenon has attracted a lot of attention due to its potential applications in memory devices. For instance, the resistive random access memory (ReRAM), based on RS effect, is a potential candidate for the next generation of memory devices.[1,2] Features such as high scalability, high speed, and low power operation give an advantage over the flash memory technology, prone to endurance issues and low speed. Structural simplicity and higher ON/OFF ratio also gives ReRAM an edge over other promising technologies like phase change RAM and spin transfer torque RAM. Binary oxides like TiO_2, NiO, ZnO, CuO, TaO_2, HfO_2, etc.[2,3] have been a popular choice for RS studies. Incorporation of magnetic components in a RS system provides an additional degree of freedom and may allow controlling the switching voltage with the external magnetic fields.

RS phenomenon has been extensively studied both experimentally and theoretically for over 40 years. Most of the existing theories of RS phenomenon are widely based on dynamic percolation model, e.g. forming and rupturing of conduction filaments[4–7] or on the migration of oxygen vacancies.[8–10] Recently, a new mechanism of RS based on self-trapped electrons and holes has been proposed.[11–13] For example, Chen *et al.* show the evidence of current injection modulation of saturation magnetization in their RS system.[11] In this work, we propose self-trapped electrons based RS mechanism in magnetic granular multilayers and the magnetic field dependence of the electron injection. We show model calculations and experiments to support this. Our calculations suggest that the switching voltage may be significantly decreased with increasing the magnetic field. We also measure *I-V* characteristics in Co/Al_2O_3 granular multilayers and demonstrate experimentally

[a]Alan Kalitsov and Ajeesh M. Sahadevan contributed equally to this work.
[b]Author to whom correspondence should be addressed. Electronic mail: eleyang@nus.edu.sg

that the switching voltage can be controlled with the external magnetic field. Our experimental observation is in good agreement with the proposed theory qualitatively.

In order to observe RS in granular multilayers it is required to apply high voltages about 10-20 V. This is so-called the forming process without which the device is an insulator. Therefore, we describe a formed granular multilayered structure by considering a simple tight-binding like model of a two-dimensional (20 by 15) cluster with classical localized moments $\boldsymbol{M}_i$ which mimic magnetic granules. A magnetic granule is represented by a single site. Since the device is formed and in conducting state, there is an overlap between the wave functions of the nearest-neighbour granules, therefore, conduction electrons can flow between the nearest-neighbour sites. We describe the transport properies of magnetic granular multilayers with a two-dimensional model because the main contribution to the current through magnetic granular multilayers comes from the first layer of magnetic granules. We model the field dependence of the directions of the localized moments with the Langevin function. The magnetic moments are randomly oriented in the absence of the external magnetic field and become aligned towards the direction of the field at high magnetic fields close to the saturation field H_s. The localized moments are coupled with conduction electrons through the local exchange interacton J. The two-dimensional granular cluster is coupled with two non-magnetic leads. The Hamiltonian of the system has the form

$$\hat{H} = \hat{H}_C + \hat{H}_L + \hat{H}_R + \hat{H}_{CL} + \hat{H}_{CR} + H.c., \tag{1}$$

where $\hat{H}_C$ is the Hamiltonian of the uncoupled cluster, $\hat{H}_{L/R}$ is the Hamiltonian of the left/right lead, and the term $\hat{H}_{CL/CR}$ describes the coupling of the granular cluster to the left/right lead. $\hat{H}_C$ has the form

$$\hat{H}_C = U \sum_{i,\sigma} \hat{c}_i^{\dagger\sigma} \hat{c}_i^{\sigma} + t \sum_{i,j,\sigma} \hat{c}_i^{\dagger\sigma} \hat{c}_j^{\sigma} - J \sum_{i,\alpha,\beta} \hat{c}_i^{\dagger\alpha} (\sigma_{\alpha\beta} \cdot M_i) \hat{c}_j^{\beta}, \tag{2}$$

where U is the electrostatic potential which is equal to $U_{0(ch)}$ if the granules are uncharged (charged), t is the spin-independent effective hopping integral between the nearest-neighbour granules, $\hat{C}_i^{\dagger\sigma}$ and $\hat{C}_i^{\sigma}$ are the creation and annihilation operators of the conduction electron with spin σ on site i, and $\sigma_{\alpha\beta}$ is the vector of Pauli matrices.

We calculate the electric current I through the system when a voltage V is applied across the device. Our calculations are based on Non-Equilibrium Green Functions formalism. The details of the approach can be found elsewhere.[14] First we diagonalize $\hat{H}_C$ and find the retarded Green Function of the uncoupled cluster g^r. Next we find the retarded Green Function of the coupled system by solving the Dyson equation

$$G^r = g^r + g^r \Sigma_L^r G^r + g^r \Sigma_R^r G^r, \tag{3}$$

where $\Sigma_{L/R}^r$ is the retarded self-energy due to connection of the cluster to the left/right lead. We assume that $\Sigma_{L/R}^r$ is independent of energy. The final expression for the charge current becomes

$$I = \frac{e}{h} \int d\varepsilon [f_L(\varepsilon)] Tr[G^a \Gamma_R G^r \Gamma_L]. \tag{4}$$

where G^a is the advanced Green Functions of the coupled system, $\Gamma_{L/R} = i(\Sigma_{L/R}^r - \Sigma_{L/R}^a)$ and $f_{L/R}$ is the Fermi-Dirac distribution functions in the left/right lead.

In the calculations we use the parameters $U_0 =$ -0.5 eV, $U_{ch} =$ -0.6 eV, $t = 0.1$ eV, $J = 0.85$ eV, and $\Gamma_L = \Gamma_R = 0.1$ eV. The ratio of J/t is an important parameter in our calculations. We choose $J/t >> 1$ because the concentration of magnetic granules in granular multilayers is small and hence the effective hopping integral (t) is small. In this case there are two well separated bands. In Fig. 1(a)–1(c) we plot the density of states of the granular system at different values of the magnetic field. The Fermi level is at 0 eV. The valence band is completely filled and the occupation of the conduction band depends on the magnetic field. It is clear from Fig. 1(b) and 1(c) that the bandwidths increase with magnetic field. When magnetic granules become charged, the shift of the electrostatic potential leads to the shift of the density of states. According to the results of Fig. 1(a)–1(c) one can expect a transition from tunneling to ohmic regime depending on the charging status at small

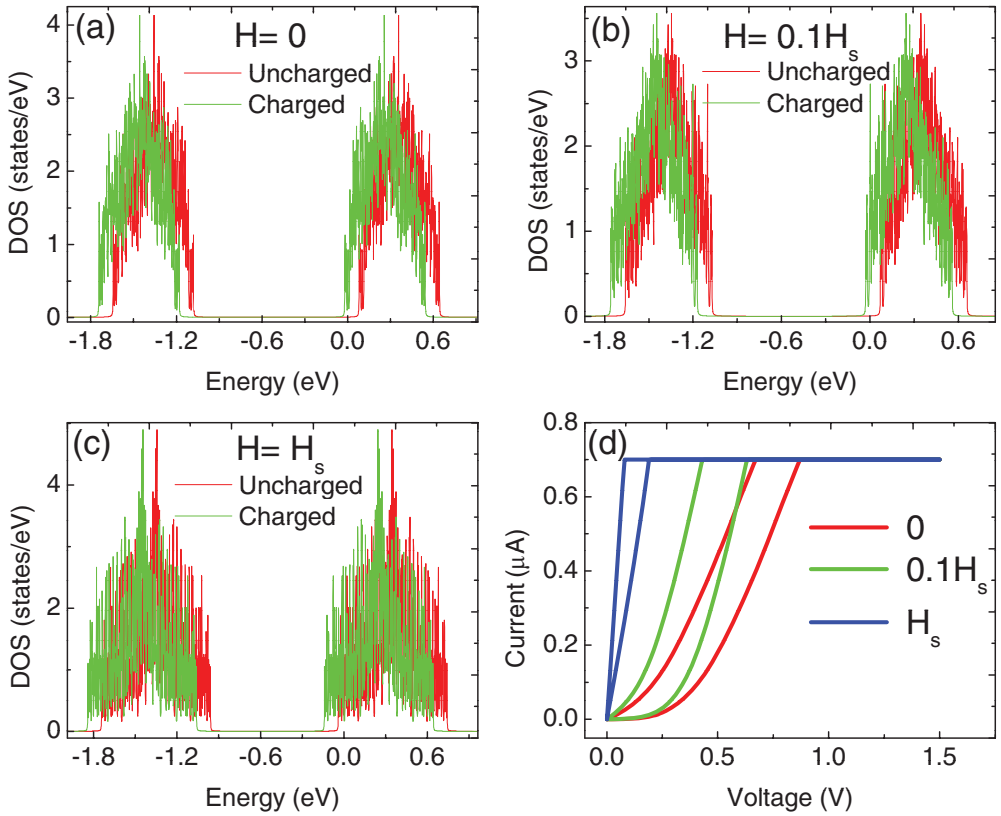

FIG. 1. Density of states (DOS) with different values of external magnetic field (H), for both uncharged and charged conditions at $H = 0$ (a), $H = 0.1H_s$ (b), and $H = H_s$ (c). The Fermi level is at 0 eV. The valence band is completely filled for all cases. After charging, the center of the conduction band moves closer to the fermi level. The occupation of the conduction band depends on the magnetic field. At $H = H_s$ the conduction band is partially filled. (d) Calculated I-V characteristics of the RS system for different H. At a fixed magnetic field the system changes from a HRS to LRS when the voltage is swept from 0 to 1.5 V. At higher magnetic field the resistance of the system reduces, and it reaches a compliance limit at a smaller applied voltage.

magnetic fields ($H = 0$ or $H = 0.1\ H_s$), while the system is in the ohmic regime at H_s regardless of the charging level.

In Fig. 1(d) we show the I-V characteristics at different values of the magnetic field. We set the compliance current, which is a parameter in our model because higher currents induce ionic motion. Taking into account ionic motion at high voltages is beyond the goal of present calculations. The switching voltage significantly decreases with increasing the magnetic field. The underlying mechanism is the influence of the magnetic field on electron occupation of the conduction band. The conduction band is almost empty without the magnetic field and the corresponding current at small voltages (< 0.1 V) is very small. When the voltage is increasing, the number of conduction electrons is increasing exponentially which leads to the rapid increase of the current. At higer voltages (> 0.5 V) the conduction band is occupied and the current linearly depends on the voltage. At higher voltage, the granules become charged and the change in electrostatic potential switches the system from HRS to LRS. As the voltage across the system is reduced, the granules retain the charge and this leads to hysteresis in the I-V curves.

The I-V characteristic at $0.1H_s$ is similar with that one at zero field but since the cluster's bandwidth gets larger, smaller voltage is needed to occupy the conduction band and the transition to the ohmic regime occurs at smaller voltage (~ 0.3 V). At the saturation magnetic field, the bandwidth is large enough and the conduction band is partially occupied even at zero voltage. The corresponding I-V characteristic at H_s indicates that the system is in the ohmic regime regardless of the charging level of granules.

The magnetic field induced transition from tunneling to ohmic regime occurs if the bottom of the conduction band in absentia magnetic field is above the Fermi level, while at high magnetic fields it lies below the Fermi level. For the two-dimensional tight-binding model the critical magnetic field H_c, at which this transition occurs, can be defined from $U + J - 4\tilde{t}(H_c) = 0$, where $\tilde{t}$ is the average effective hopping integral between the nearest-neighbour granules. The value of $\tilde{t}$ depends on the applied magnetic field. If we assume that the effective hopping integral between the nearest neighbor granules i and j, t_{ij} is proportional to the tunneling probability of the conduction electron to tunnel between granules i and j, and the spin polarization of granules is 100%, then the electron hopping can be written as $t_{ij} = t(1 + \cos\theta_{ij})/2$, where θ_{ij} is the angle between the magnetic moments of granules i and j[15] At the saturation field magnetic moments of all granules are aligned towards the direction of the field and $\tilde{t}(H_s) = t$. In the absence of an external magnetic field the magnetic moments of granules are randomly oriented and $\tilde{t}(0) = t/2$.

The value of $U + J$and t are important parameters in our calculations. The system is always in the tunneling regime if $U + J > 4t$, and the system is always in the ohmic regime for $-2t < U + J < 2t$. Experimentally it is possible to control $U + J$ parameter by choosing different oxides (different U) and different magnetic material of granules (different J). The t parameter can be controlled by fabricating granular multilayers with different concentration of magnetic granules. Larger value of t corresponds to granular multilayers with higher granular concentration.

We fabricate Co/Al_2O_3 magnetic granular system in order to study experimentally the magnetic field dependence of resistive switching behavior in magnetic granular system. The inset in Fig. 2(a) shows a schematic of the system used in this study. Thermally evaporated Cr (5 nm)/Au (45 nm) bottom-layer are deposited on Si (100) substrates with a 400 nm thick thermally oxidized SiO_2 layer. Ten layers of [Al^2O^3 (4 nm)/Co (0.5 nm)] are subsequently deposited and the structure is capped by a 4 nm Al_2O_3 layer. Co and Al_2O_3 are deposited in an ultra-high vacuum (10^{-9} torr) magnetron sputtering chamber using dc and rf sources, respectively. Electrical contact pads (80 μm $\times$ 80 μm) are formed by thermally evaporated Cr (5 nm)/Au (45 nm) and the distance between two adjacent contacts for measurements is 50 μm. Figure 2(a) shows the cross sectional transmission electron microscope (TEM) image of Co/Al_2O_3 granular multilayers. Co nanoparticles are clearly visible, embedded in an Al_2O_3 insulating matrix. Co forms a discontinuous layer of nanodots with a diameter considerably larger than the nominal layer thickness of 0.5 nm.[16,17] X-ray photoelectron spectroscopy (XPS) depth-profile analysis shows alternate peaks of [Al (2p), O (1s)] and Co (2p).

The *I-V* measurements have been carried out at room temperature in ambient conditions. The initial state of the Co/Al_2O_3 is an insulator. Without the forming process, switching behavior is not observed between 0 and 2 V range. Forming process is necessary for most RS devices to show reproducible switching behavior.[1,18] The forming voltage for the system is around 12 V as shown in Fig. 2(b), with a current compliance of 35 mA. Appling a high voltage induces ionic motion, leading to non-reversible structural changes in oxides and making granular multilayers conductive. The current compliance is required to prevent the device from non-reversible ionic motion. Immediately after the forming process, the *I-V* characteristics are obtained in the voltage range of 0 - 2 V, keeping the current compliance at 35 mA. The range of measured currents is larger than the theoretical range because of the larger size of the electrodes. The system shows a typical characteristic of threshold RS as shown in Fig. 2(c). It would be interesting to mention that only a small part of the structure may be contributing to the switching process as has been reported in similar systems where electrons flow between nanodots separated by insulating barriers.[17,19,20]

The effect of magnetic field on the switching behavior is studied by applying the magnetic field parallel to the direction of the average current in the film plane. It is clear from Fig. 2(d) that the switching voltage (V_t), at which the resistance state changes from a HRS to a LRS, decreases with an increase in magnetic field. For example V_t decreases from 0.83 V at zero field to 0.42 V at 52 mT as shown in the inset of Fig. 2(d). This field dependent modulation of the junction current has never been observed in the magnetic granular system, but reported in 83 nm NiO fim.[21] With the presence of Ni rich centers (oxygen vacancies), these Ni rich regions work as magnetic granules in an oxide environment of NiO and the proposed theory may be used to explain the observed magnetic field dependence.

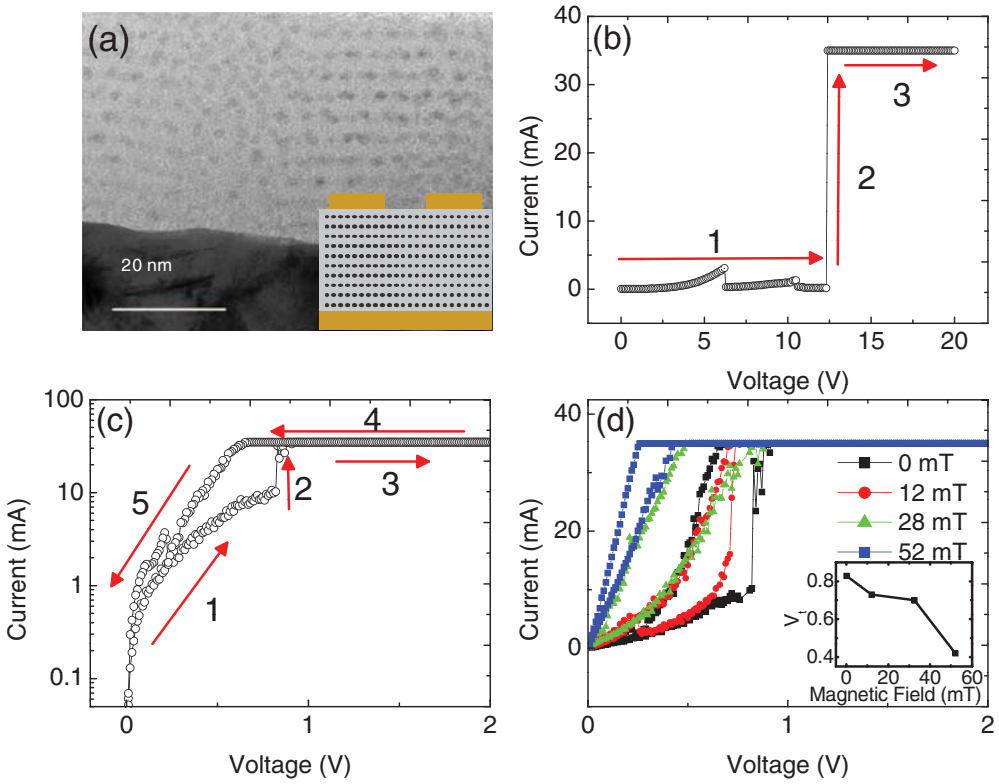

FIG. 2. (a) Cross sectional transmission electron microscope (TEM) image of Co/Al_2O_3 multilayers. The dark spots are Co islands and the white region is an Al_2O_3 insulating matrix. The Al_2O_3 thickness is 4 nm and the nominal thickness of a Co layer is about 0.5 nm. The inset shows a schematic of Co/Al_2O_3 multilayer system. (b) *I-V* characteristics of the device during the forming process. The forming voltage is around 12 V. (c) Threshold resistive switching behavior due to charge accumulation in the granules. (d) Experimental *I-V* characteristics of threshold switching for different external magnetic fields. The inset shows how the switching voltage (V_t) changes with the external magnetic fields.

In conclusion, we propose a theoretical model of magnetic field dependence of threshold resistive switching in magnetic granular system based on the self-trapped electrons mechanism. We show that the switching voltage can be significantly decreased with increasing the magnetic field for a certain range of model parameters. We qualitatively support our theoretical calculations by measuring the magnetic field dependence of threshold resistive switching behavior in Co/Al_2O_3 granular multilayers and demonstrate experimentally an ability to control the switching voltage with external magnetic field.

ACKNOWLEDGMENTS

This work was partially supported by the Singapore Ministry of Education Academic Research Fund Tier 2 (MOE2008-T2-1-105) and National Research Foundation under CRP Award No. NRF-CRP 4-2008-06.

[1] R. Waser and M. Aono, Nat. Mater. **6**, 833 (2007).
[2] H. Akinaga and H. Shima, Proc. IEEE **98**, 2237 (2010).
[3] A. Sawa, Mater. Today **11**, 28 (2008).
[4] J. S. Lee, S. B. Lee, S. H. Chang, L. G. Gao, B. S. Kang, M. J. Lee, C. J. Kim, T. W. Noh, and B. Kahng, Phys. Rev. Lett. **105**, 205701 (2010).
[5] K. M. Kim, B. J. Choi, and C. S. Hwang, Appl. Phys. Lett. **90**, 242906 (2007).
[6] G. S. Park, X. S. Li, D. C. Kim, R. J. Jung, M. J. Lee, and S. Seo, Appl. Phys. Lett. **91**, 222103 (2007).
[7] D. H. Kwon, K. M. Kim, J. H. Jang, J. M. Jeon, M. H. Lee, G. H. Kim, X. S. Li, G. S. Park, B. Lee, S. Han, M. Kim, and C. S. Hwang, Nat. Nanotechnol. **5**, 148 (2010).

[8] M. Quintero, P. Levy, A. G. Leyva, and M. J. Rozenberg, Phys. Rev. Lett. **98**, 116601 (2007).
[9] M. J. Rozenberg, M. J. Sanchez, R. Weht, C. Acha, F. Gomez-Marlasca, and P. Levy, Phys. Rev. B **81**, 115101 (2010).
[10] J. H. Hur, M. J. Lee, C. B. Lee, Y. B. Kim, and C. J. Kim, Phys. Rev. B **82**, 155321 (2010).
[11] Y. S. Chen, G. J. Lian, G. C. Xiong, and T. Venkatesan, Appl. Phys. Lett. **98**, 232513 (2011).
[12] Y. S. Chen, L. P. Chen, G. J. Lian, and G. C. Xiong, J. Appl. Phys. **106**, 023708 (2009).
[13] D. M. Ramo, A. L. Shluger, J. L. Gavartin, and G. Bersuker, Phys. Rev. Lett. **99**, 155504 (2007).
[14] Y. Meir and N. S. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).
[15] J. Inoue and S. Maekawa, Phys. Rev. B **53**, 11927 (1996).
[16] R. Brucas, M. Hanson, P. Apell, P. Nordblad, R. Gunnarsson, and B. Hjorvarsson, Phys. Rev. B **81**, 224437 (2010).
[17] H. Yang, S.-H. Yang, and S. S. P. Parkin, Nano Lett. **8**, 340 (2008).
[18] S. 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).
[19] C. Gould, A. Slobodskyy, D. Supp, T. Slobodskyy, P. Grabs, P. Hawrylak, F. Qu, G. Schmidt, and L. W. Molenkamp, Phys. Rev. Lett. **97**, 017202 (2006).
[20] D. G. Austing, S. Tarucha, P. C. Main, M. Henini, S. T. Stoddart, and L. Eaves, Appl. Phys. Lett. **75**, 671 (1999).
[21] S. Das, S. Majumdar, and S. Giri, J. Phys. Chem. C **114**, 6671 (2010).