Reproducing resistive switching effect by Soret and Fick diffusion in resistive random access memory (ReRAM)

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Abstract

It is widely received that resistive switching of resistive random access memory (ReRAM) is caused by formation and rupture of conductive filament consisting of oxygen vacancies, $V_O$'s. However, driving forces that migrate oxygen vacancies are not elucidated yet.

In this paper, reset was attempted by injecting saw-tooth shaped voltage pulses with various rise time, $t_{\text{lead}}$, to a Pt/NiO/Pt cell in a low resistance state. Reset occurred for long $t_{\text{lead}}$, whereas the resistance was rather reduced for short $t_{\text{lead}}$. The latter cannot be explained by generally accepted switching models that assume Fick diffusion and electric field drift. Therefore, we re-examined driving forces and succeeded in reproducing pulse response data for wide $t_{\text{lead}}$ range by simulating $V_O$ migration assuming Fick and Soret diffusion.

It was suggested that the change of resistance is decided as a result of the competition of Fick and Soret diffusion.

1. Introduction

A filament model, in which a conductive filament (CF) consisting of oxygen vacancies ($V_O$’s) is formed in the metal oxide (MO) film of an EL/MO/EL structure by a forming process and the resistive switching effect is caused by generation and repair of oxygen vacancies, is widely accepted, where EL and MO represent electrode and metal oxide, respectively.$^1$ However, driving forces of $V_O$ migration that cause reset and set switching are still unclear. Koh et al. reported that the oxygen reservoir of a Pt/NiO/Pt structure is the NiO film itself surrounding the CF.$^2$ Therefore, a driving force that is different from an electric field drift that is perpendicular to the EL interface is required to cause set switching. One of the candidates for this driving force is the Soret force, which works in the direction of the temperature, $T$, gradient.$^3$

In this paper, we examined Soret diffusion as a driving force instead of electric field drift of $V_O$’s. We succeeded in reproducing pulse response data for wide rise time, $t_{\text{lead}}$, range by simulating $V_O$ migration assuming Fick and Soret diffusion as driving forces. The change of resistance is suggested to be decided by the relative magnitude of Fick and Soret diffusion.

2. Experiment

Reset was attempted for Pt(100nm)/NiO(60nm)/Pt (100nm) cells in a low resistance state (LRS) that have resistance, $R_{\text{LRS}}$, of 70-120 $\Omega$ by injecting a saw-tooth shaped voltage pulse with the rise time of $t_{\text{lead}}$ as shown in the inset of Fig. 1(a). The pulse height, $V_p$, was increased from 0.3 to the $V_p$ at which reset switching or reduction of $R_{\text{LRS}}$ was confirmed in steps of 0.1 V.

Reset was attempted by simulating $V_O$ migration using commercial software (COMSOL Multiphysics). We assumed Soret and Fick diffusion, whereas electric field drift and Fick diffusion are generally adopted as driving forces of $V_O$ migration.

3. Results and Discussion

Figs. 1(a) and (b) show typical current responses to injected saw-tooth shaped voltage waves with $t_{\text{lead}}$ of 3 ms and 30 $\mu$s, respectively. Reset did not occur for $t_{\text{lead}}$ of 30 $\mu$s and the $R_{\text{LRS}}$ was reduced to the value smaller than 0.9 times the initial $R_{\text{LRS}}$ value at $V_p$ of 0.9 V, whereas reset occurred at $V_p$ of 0.8 V for $t_{\text{lead}}$ of 3 ms. Fig. 2 shows $V_p$ dependence of resistances for $t_{\text{lead}}$ of 300 ms, 3 ms, and 30 $\mu$s.

The occurrence of reset was confirmed for $t_{\text{lead}}$ of 300 ms and 3 ms, whereas $R_{\text{LRS}}$ was reduced for $t_{\text{lead}}$ of 30 $\mu$s.

Reset switching was attempted by the simulation in which Fick ($\propto dV_O/dr$) and Soret ($\propto dT/dr$) diffusion are assumed to be driving forces for $V_O$ migration. Fluxes of $V_O$'s for Fick and Soret diffusion, $J_{\text{Fick}}$ and $J_{\text{Soret}}$, are as follows:

$$J_{\text{Fick}} = D_{\text{Fick}} n_{\text{Fick}} \frac{dV_O}{dr}, \quad (1)$$

$$J_{\text{Soret}} = -D_{\text{Fick}} n_{\text{Soret}} \frac{dT}{dr}, \quad (2)$$

where $D_{\text{Fick}}$, $n_{\text{Fick}}$, and $n_{\text{Soret}}$ are the diffusion constant, the density, and the Soret coefficient of $V_O$’s as a function of the radial coordinate, $r$, respectively, assuming a cylindrical cell structure (Fig. 3(a)). The electric conductivity of CF and the NiO film is assumed to be $\sigma = \sigma_0 \exp(-E_c/k_B T)$, where $\sigma_0$ and $E_c$ as a function of $n_{\text{Fick}}$ were given by Figs. 3(b) and (c), respectively, and $k_B$ is Boltzmann constant.

The resistance increased with increasing the voltage when saw-tooth wave with $t_{\text{lead}}$ and $V_p$ of 1.80 ms and 0.72 V, respectively, was injected (Fig. 4(a)). On the other hand, the resistance decreased by 7% after the application of saw-tooth wave with $t_{\text{lead}}$ and $V_p$ of 1.80 ms and 1.02 V, respectively (Fig. 5(a)). $V_O$’s consisting of the CF receive the force by Eq. 1 toward the outside of the CF because $n_{\text{Fick}}$ in the CF is higher than that of the outside of CF. On the other hand, $V_O$’s receive the force toward a current path that is a heating center owing to
Joule heating from the surrounding area because \( T \) in the CF is higher than that of the outside of CF (Fig. 6(a)). Therefore, when \( t_{\text{lead}} \) is long enough to maintain thermal equilibrium continuously, \( J_{\text{Soret}} \) is small because \( dT/dr \) in Eq. 2 becomes small (black line in Fig. 6(b)) and the occurrence of reset is confirmed (A \( \Rightarrow \) C in Fig. 4(b)). However, when \( t_{\text{lead}} \) is short so that thermal equilibrium cannot be maintained, the shorter the \( t_{\text{lead}} \) becomes, the larger \( dT/dr \) becomes (red line in Fig. 6(b)), making \( J_{\text{Soret}} \) dominant and reset switching is prevented (D \( \Rightarrow \) F in Fig. 5(b)).

4. Conclusions
Experimentally obtained pulse response property can be reproduced successfully by the simulation that involves Soret and Fick diffusion without assuming electric field drift.

References
1) M. J. Lee et al., Nano Lett. 9, 1476 (2009).