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## Effect of oxygen content of the LaAlO<sub>3</sub> layer on the synaptic behavior of Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristors for neuromorphic applications

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### ABSTRACT

We report the effect of the oxygen content of the LaAlO<sub>3</sub> layer on the synaptic behavior in the Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristor for neuromorphic applications. As the oxygen-content decreases, the current becomes larger and the spike time-dependent plasticity (STDP) becomes less sensitive to the time difference between pre- and post-synaptic spike voltage. In addition, the conduction mechanism, which was found to be a combination of thermionic and Poole-Frenkel emissions, and the effect of oxygen content are explained in association with the oxygen vacancy in the LaAlO<sub>3</sub> layer. The trade-off between large current and efficient STDP can be controlled by the oxygen content. Furthermore, the results of extracting the synaptic strength-based model parameters indicate that the Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> shows the efficient STDP characteristics in comparison to previously reported memristor materials.

### 1. Introduction

With regard to a diverse oxide-based combined structure, the heterostructures of perovskite oxides are emerging as one of the most fascinating materials with a broad spectrum of functional properties such as two-dimensional electron gas, orbital reconstruction, interfacial superconductivity, ferromagnetism, charge writing, resistive switching (RS), giant thermoelectric effect, and colossal ionic conductivity [1–3]. In particular, the high-mobility characteristics at the interface between two perovskite materials have been frequently observed via intensive experimental and theoretical investigations on the physics such as built-in electric field [4], oxygen vacancy (V<sub>O</sub>) migration [5], and cation intermixing [6]. Thus, many researchers have focused bulky and/or interfacial characteristics of the perovskite materials due to various functional features. Applications based on these characteristics have been reported such as the transparent devices using the wide bandgap [7], the nonvolatile memory with the capacitor-like structure [8], and the sensor of the gas responding the palladium nanoparticles [9].

On the other hand, neuromorphic computing to mimic the brain system has been studied to verify low power and high speed device beyond Von Neumann computing system by many researchers [10–12]. In particular, they have developed the device to emulate the neuromorphic computing system where the synapse is a structure that passes an electrical or chemical signal between neuron pair in the nervous

system. The one of the devices to emulate these neuromorphic systems is the memristor which enables the bias/time-dependent gradual resistive switching (RS) and is composed of simple metal/insulator/metal structure such as Pr<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub> [13], Ta<sub>2</sub>O<sub>5</sub> [14], γ-Fe<sub>2</sub>O<sub>3</sub> [15], amorphous InGaZnO [11,16], and perovskite oxide such as LaAlO<sub>3</sub>/SrTiO<sub>3</sub>, BiFeO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub>, and amorphous SrTiO<sub>3</sub> [7,8,17,18].

Therefore, the synaptic behavior of perovskite materials-based memristor needs to be investigated and assessed in viewpoint of the feasibility of neuromorphic system. In real, perovskite materials-based memristors have attracted much attention as the RS devices. As previously reported, perovskite oxide materials-based memristors showed the RS characteristics between a low resistance state (LRS) and a high resistance state (HRS) associated with the generation of oxygen vacancies (V<sub>O</sub>s) which is based on the transition from Ti<sup>4+</sup> to Ti<sup>3+</sup> in the SrTiO<sub>3</sub> bulk [17].

In this work, we investigate the synaptic behavior in Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristor. First of all, we reveal the conduction mechanism of the Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristor by modifying the current-voltage (I-V) characteristics. Secondly, we demonstrate the spike time-dependent plasticity (STDP) features to mimic the brain system and analyze the effect of the oxygen-content in the LaAlO<sub>3</sub> layer on STDP. Finally, the STDP characteristics are quantized and compared with the previously reported results.

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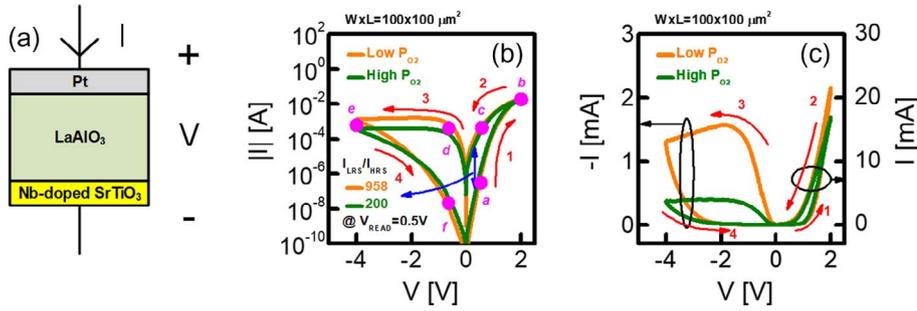


Fig. 1. (a) Device structure and  $P_{O_2}$ -dependent RS I-V behaviors of (b) log and (c) linear scale in the Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristor.

## 2. Device fabrication and measurement setup

The Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristors were fabricated as shown in Fig. 1(a). The Nb-doped SrTiO<sub>3</sub> was prepared as the conductive substrate [19,20], which plays the role of bottom electrode (BE). Then, the LaAlO<sub>3</sub> was grown as the RS layer by the molecular beam epitaxy at 640 °C on the TiO<sub>2</sub>-terminated Nb-doped SrTiO<sub>3</sub> substrate. It is known that the Nb-doped SrTiO<sub>3</sub> is a very good substrate due to a small lattice mismatch with LaAlO<sub>3</sub>. It is very useful when one characterizes physical properties of the epitaxial LaAlO<sub>3</sub> in the out-of-plane direction. The oxygen-poor and oxygen-rich samples were fabricated by varying the oxygen partial pressure ( $P_{O_2}$ ) from  $10^{-6}$  and  $10^{-4}$  Torr, respectively. The  $P_{O_2}$  controls the concentration of  $V_{OS}$  in RS film during the LaAlO<sub>3</sub> growth. The thickness of LaAlO<sub>3</sub> is 7.726 nm, which corresponds to the 20 unit cells. Finally, the 100 nm-thick Pt was deposited as top electrode (TE) with the area of  $100 \times 100 \mu\text{m}^2$  by using the dc magnetron sputtering with the power of 50 W.

In order to analyze the electrical characteristics, the dc current-voltage (I-V) and potentiation/depression pulse characteristics were measured at a room temperature and dark condition with Keithley-4200 semiconductor characterization system.

## 3. Bipolar resistive switching and memristive characteristics

The RS I-V behaviors of the Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristors are shown in Fig. 1(b) and (c). The sequence of applying voltage is  $0\text{ V} \rightarrow +2\text{ V}$  (SET process)  $\rightarrow 0\text{ V} \rightarrow -4\text{ V}$  (RESET process)  $\rightarrow 0\text{ V}$ , where both the polarity of voltage and the direction of current flow are shown in Fig. 1(a). The SET process is the RS from a high-resistance state (HRS) to a low-resistance state (LRS) and the RESET process is the RS from LRS to HRS. The I-V hysteresis loop in Fig. 1(b) and (c) shows a stable bipolar RS characteristic. In Fig. 1(b), the point from *a* to *f* indicates a specific operation point. The current ratio of LRS to HRS ( $I_{LRS}/I_{HRS}$ ) at read voltage ( $V_{\text{READ}} = 0.5\text{ V}$ ) is 958 and 200 in the oxygen-poor and oxygen-rich sample, respectively. The voltage polarity of SET/RESET is opposite to previous work [7], where  $V_{OS}$  are supplied from the SrTiO<sub>3</sub> bulk, because in our case the initial density of  $V_{OS}$  is quite high due to relatively low  $P_{O_2}$  during the LaAlO<sub>3</sub> deposition. Consistently, it was found by the XPS analysis that the oxygen concentration in LaAlO<sub>3</sub> can be controlled by modulating the oxygen partial pressure [21].

The conduction mechanism is somewhat complicated and needs to be fully understood especially in memristive devices because it is dependent on a history of I-V operation. Fig. 2 elucidates the transport mechanism and illustrates it at individual operation point (*a-f*). For HRS at a low  $V_{\text{READ}}$  (the voltage conditions lower than *a* in Fig. 2(a) and lower than *f* in Fig. 2(c)), the conduction follows the thermionic emission:

$$I = AA^*T^2 \exp\left(-\frac{q\phi_B}{kT}\right) \exp\left(\frac{qV}{\eta kT}\right) \quad (1)$$

where the  $I$  is the current,  $A$  is the area of device,  $A^*$  is the Richardson constant,  $kT$  is the thermal energy,  $\phi_B$  is the effective barrier height,  $q$  is

the electron charge, and  $\eta$  is the ideality factor. The extracted  $\phi_B$  is 0.77 eV and 0.72 eV for the oxygen-poor and oxygen-rich devices, respectively.

For either the HRS at a high  $V_{\text{READ}}$  (the voltage conditions higher than *a* in Fig. 2(b) and higher than *f* in Fig. 2(d)) or the LRS at a higher/lower  $V_{\text{READ}}$  than *c/d* in Fig. 2(b)/(d), the conduction is dominated by the Poole-Frenkel (P-F) emission:

$$I = AqN_C\mu E \exp\left(-\frac{q\phi_B}{kT}\right) \exp\left(\frac{q}{\eta kT} \sqrt{\frac{qE}{\pi\epsilon}}\right) \quad (2)$$

where  $N_C$  is the density of states in the conduction band,  $\mu$  is the carrier mobility,  $\epsilon$  is the permittivity of the dielectric constant, and  $E$  is the electric field. The P-F emission is a means by which electrons with enough energies that can overcome the energy barrier can escape the localized states after the electrons are generally trapped in the localized states, in other words, the P-F mechanism is the continuative hopping phenomenon. The  $\phi_B$  for P-F emission is extracted to 0.5 eV and 0.56 eV for the oxygen-poor and oxygen-rich samples, respectively. According to Mitra et al. [22],  $V_{OS}$  in the LaAlO<sub>3</sub> can be formed within a variety of ranges of charge state and energy level, such as neutral  $V_{O}^0$  ( $E_C - 2.19\text{ eV}$ ),  $V_{O}^+$  ( $E_C - 1.44\text{ eV}$ ), and  $V_{O}^{2+}$  ( $E_C - 0.62\text{ eV}$ ). In particular, since the  $V_{OS}$  near the  $E_C$  of the LaAlO<sub>3</sub> layer, which determine the rate of electron migration, have positive charge, the energy barrier lowering due to the positively charged  $V_{OS}$  may occur. Therefore, lower  $\phi_B$  in the case of higher density of  $V_{OS}$  (oxygen-poor sample) suggests that the RS characteristic in Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristor results from the hopping and migration via  $V_{OS}$  in the LaAlO<sub>3</sub> layer. Lower  $\phi_B$  in the case of higher density of  $V_{OS}$  (oxygen-poor sample) suggests that the RS characteristic in Pt/LaAlO<sub>3</sub>/Nb-doped SrTiO<sub>3</sub> memristor results from the hopping and migration via  $V_{OS}$  in the LaAlO<sub>3</sub> layer.

The memristive behavior is then characterized by the potentiation/depression pulse measurement with the duration of SET/RESET pulse ( $t_{\text{SET}}/t_{\text{RESET}} = 1\text{ ms}$ , the SET/RESET voltage ( $V_{\text{SET}}/V_{\text{RESET}} = +2\text{ V}/-2\text{ V}$ , and 50 cycles. The read current ( $I_{\text{READ}}$ ) is sampled during the read period ( $V_{\text{READ}} = 0.5\text{ V}$ ) which is inserted between adjacent potentiation/depression pulse trails (Fig. 3(a)). The measured  $I_{\text{READ}}$  with the increase of the number of cycles is observed as shown in Fig. 3(b). In comparison with the oxygen-rich sample, the oxygen-poor sample shows a higher current and larger difference between the potentiation and depression. Our results suggest that the higher  $V_{O}$  density (oxygen-poor condition) is advantageous in terms of the memristive viewpoint because not only the magnitude of current but also the change of current for the potentiation and depression is very important to obtain the properties of analog memory associated with synaptic behavior and improve the operating speed.

## 4. Spike time-dependent plasticity characteristics for neuromorphic applications

Nervous system between the axon and dendrite follows the role of memory in the following mechanisms. The ends of the axon and the dendrite are applied to the pre- and post-synaptic spike ( $V_{\text{pre}}$  and  $V_{\text{post}}$ ),

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