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Resistance state-dependent barrier inhomogeneity and transport mechanisms in resistive-switching Pt/$SrTiO_3$ junctions

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We investigated the current–voltage ($I-V$) and photocurrent characteristics of Pt/Nb-doped $SrTiO_3$ (001) single-crystal junctions that exhibit resistive-switching behaviors. The temperature-dependent $I-V$ data and the photocurrent spectra showed that the barrier height fluctuation depended on the resistance state but the mean barrier height was nearly constant regardless of the junctions' resistance state. In addition, local barrier height variations allowed transitions from thermionic to tunneling transport for the low-resistance state. © *2011 American Institute of Physics*.
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Resistive switching (RS) phenomena in metal/oxide/metal structures have gained increasing attention due to their potential for next-generation nonvolatile memory devices.[1–11] It is thought that the migration of oxygen vacancies plays a key role in bistable resistance states. The concentration and spatial distribution of oxygen vacancies (i.e., mobile dopants) can change bulk resistivity and interface potential profiles,[2–7] and also induce compositional variation, resulting in metallic phase formation and local Joule heating.[10,11] Thus, the RS behaviors of specific devices are very complicated and diverse. Single-crystalline metal-oxide-based rectifying junctions can exhibit RS during reversal of the applied voltage polarity, which can be model systems for investigating interfacial effects on RS characteristics.[7–9]

In the single-crystalline junctions, there is still debate as to whether the barrier height undergoes a change during RS.[5–9] Shang *et al.*[8] and Fujii *et al.*[9] reported that the Schottky barrier height (SBH) was almost invariant and trap-assisted tunneling affected the resistance states of the junctions. In contrast, based on first-principles calculations, Jeon *et al.*[5] and Tamura *et al.*[6] proposed that SBH could be varied by atomic-scale movements of the oxygen vacancies at the metal/oxide interface. To resolve this discrepancy, careful examinations of the barrier height and the transport mechanism of the resistive-switching junctions are required. In this study, we fabricated Pt/Nb-doped $SrTiO_3$ (Nb:STO) single-crystal junctions and investigated their electrical properties.

We prepared Pt electrodes on Nb:STO (001) single crystals with a doping ratio of 0.05 wt % (CrysTec, GmbH, Germany) using a radio frequency sputtering system.[7] The thicknesses of the Pt electrodes (diameter: 200 μm) were 50 and 10 nm for conventional transport and photocurrent measurements, respectively. The optical transmittance of the 10 nm thick Pt thin films was 16%–20% in the photon energy range from 1.6 to 2.2 eV, which allowed sufficient photocurrent for measurements. Then, we applied In contacts to both the bottom and side of the Nb:STO samples for Ohmic contact. The current-voltage ($I-V$) characteristics were investigated using a HP 4156B semiconductor parameter analyzer. The photocurrent spectra were obtained using a 200 W Xe lamp and a grating monochromator. SBH can be determined from the threshold photon energy for the photocurrent; this technique is referred to as internal photoemission spectroscopy (IPES).[12,13] Compared with conventional $I-V$ and capacitance–voltage measurements, IPES has advantages such as insensitiveness to local potential fluctuations at the interface and no concern about electric-field-induced effects.[7,12]

Figure 1 shows the rectifying $I-V$ characteristics of the Pt/Nb:STO junction in the temperature range of 297–393 K. The current direction from the top electrode was defined as positive and the opposite direction as negative. The current values were measured while sweeping the voltage from -2 to $+2$ V and back to -2 V. The switching from high resistance state (HRS) to low-resistance state (LRS) occurs during the positive bias scan and the reverse switching occurs during the negative bias scan. The $I-V$ curves were nearly invariant for several hundreds of voltage scans. This suggests that our junctions should not undergo irreversible changes in the microstructure and the chemical composition during the repetitive RS cycles.

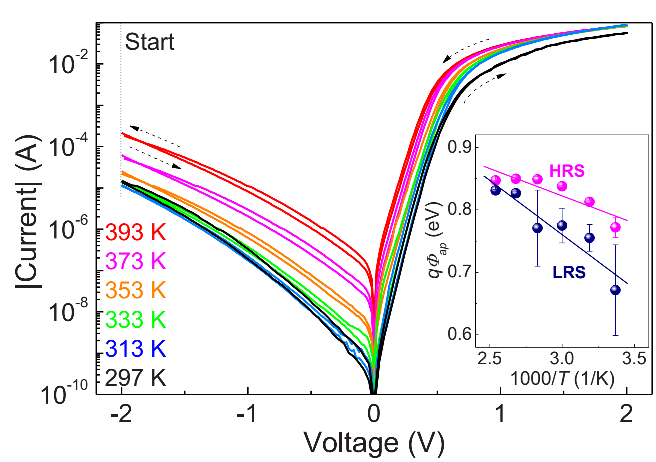

FIG. 1. (Color online) $I-V$ characteristics of a Pt/Nb:STO junction in the temperature range of 297–393 K. The inset shows a plot of apparent SBH vs $1000/T$.

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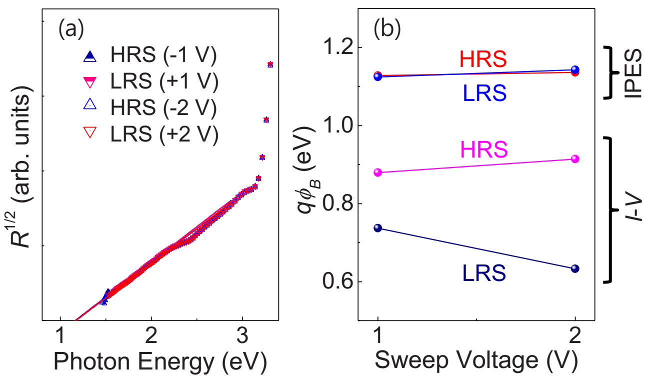

FIG. 2. (Color online) (a) IPES spectra and (b) the SBHs, estimated from IPES and $I-V$, for the Pt/Nb:STO junction with different resistance states obtained by varying dc voltage sweep polarity and range.

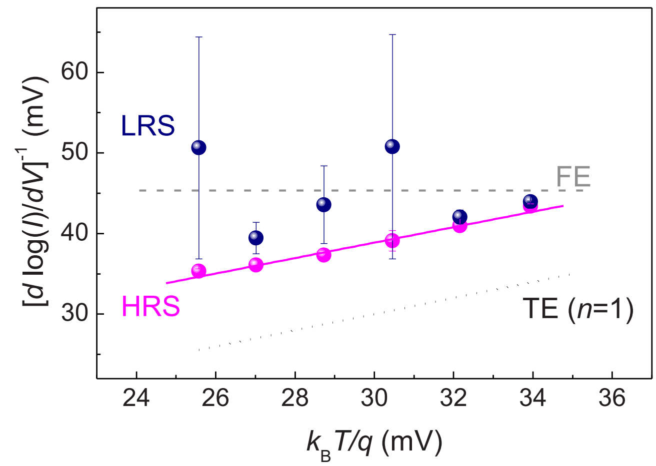

FIG. 3. (Color online) $[d\log(I)/dV]^{-1}$ vs k_BT/q curves of the Pt/Nb:STO junction for HRS and LRS.

Inset of Fig. 1 shows the SBH (Φ_{ap}) values extracted from the $I-V$ curves based on the thermionic emission (TE) model as a function of inverse temperature $(1/T)$. The Φ_{ap} values for LRS were smaller than those for HRS, as expected from the $I-V$ curves. Variation in Φ_{ap} in the measured temperature range was very large and could not be explained by the conventional Schottky diode model, which assumes a homogeneous interface barrier.[13,14] Inset of Fig. 1 shows that there was a nearly linear relationship between Φ_{ap} and $1/T$. The temperature dependence of Φ_{ap} could be described in terms of the mean barrier height $(\Phi_{B,m})$ and the standard deviation of the interface potential (σ), as follows:[14]

$$\Phi_{ap}(T) = \Phi_{B,m} - q\sigma^2/2k_BT. \tag{1}$$

The σ value for HRS (0.123 eV) was smaller than that for LRS (0.173 eV), suggesting that the local SBH was modified during RS. The barrier inhomogeneity at the metal/semiconductor junction could have been caused by intrinsic surface reconstruction, defects/doping inhomogeneity of the semiconductor surface, and so on.[7,14] It should be noted that reversing the applied voltage polarity affected the σ values. Thus, some charged defects, such as oxygen vacancies, may have contributed to the local potential profile modification at the Pt/Nb:STO interface.[5,6]

Figure 2(a) shows IPES results for the junctions with different resistance states. The photoresponsivity, R, is defined as the photoexcited electrons per incident photon. The spectral response follows the relationship $R^{1/2} \propto (h\nu - q\Phi_B)$ for $q\Phi_B < h\nu < E_g$ ($h\nu$: incident photon energy, E_g: band gap energy, and $q\Phi_B$: barrier height).[12,13] Thus, the intercept of the horizontal axis corresponds to the $q\Phi_B$ of our junction. The photocurrent spectra were obtained from an identical junction after applying dc voltage sweeps (from 0 to +1/+2 V for LRS or from 0 to $-1/-2$ V for HRS). Larger voltage sweeps caused larger resistance differences; the change in the resistance ratio was 45 and 579 for ± 1 and ± 2 V (reading voltage: 0.25 V), respectively. The SBHs estimated from IPES were nearly equal, although those from the $I-V$ measurements differed depending on the resistance state, as shown in Fig. 2(b). Our junctions can be thought of as parallel-connected nonidentical diodes due to the aforementioned inhomogeneity. Electric current will predominantly flow through the region with lower barrier height than the neighboring regions. Thus, the SBHs from the $I-V$ analyses yielded the local minima of the SBH. In contrast, the IPES provided the mean barrier height. Thus, the SBH from IPES was larger than that from $I-V$.[13,14] Therefore, the results in Figs. 2(a) and 2(b) show that RS did not influence the mean SBH but did alter the interface potential fluctuation.

Figure 3 shows $[d\log(I)/dV]^{-1}$ versus k_BT/q curves for HRS and LRS. If TE dominates the transport in metal/semiconductor junctions, the forward current is given by $J_F^{\mathrm{TE}} \propto \exp(qV/nk_BT)$, where n is the ideality factor. When field emission (FE) is dominant, electrons near the Fermi level tunnel through the barrier, and the forward current is described as $J_F^{\mathrm{FE}} \propto \exp(qV/E_{00})$, where $E_{00} = q\hbar/2(N/m^*\varepsilon_s)^{1/2}$ (m^*: effective mass of an electron and ε_s: dielectric constant of the semiconductor). The values of $[d\log(I)/dV]^{-1}$ for HRS were proportional to k_BT/q, suggesting that the TE process dominated the transport.[14] Unlike HRS, LRS exhibited no clear temperature dependence, indicating that tunneling transport occurred.[14] The combined results revealed that transition of the transport mechanism can occur depending on the resistance state.

Shang *et al.*[8] reported significant transport behavior in their RS Au/Nb:STO junctions: (1) the log(I) versus V curves measured at different temperatures were parallel in the forward bias and (2) the reverse current switched from increasing to decreasing with raising temperature as the reverse bias was increased. Based on their results, they suggested that there is a close relationship between RS and electron tunneling. Fujii *et al.*[9] also found that tunneling governed the conduction of their $SrRuO_3$/Nb:STO junctions. They suggested that RS in their junctions originated from the change in conductance through the tunneling paths rather than a change in the barrier potential profile. However, our results show that tunneling may not be a prerequisite for RS.

Based on all our experimental results, we propose illustrations of the current transport across the Pt/Nb:STO junctions as shown in Fig. 4. The SBHs estimated from the IPES showed no difference between HRS and LRS, indicating that the resistance state did not affect the mean barrier height. The $I-V-T$ measurements revealed that the spatial fluctuation of the SBHs for LRS was larger than that for HRS. In the case of LRS, certain regions with sufficiently low barriers may have allowed tunneling conduction. Such lowering of local barriers can explain both the RS and resistance state-

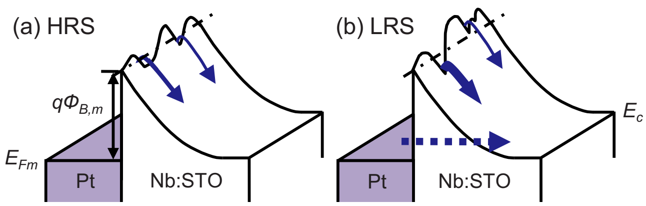

FIG. 4. (Color online) Schematic diagrams of the current transport across the Pt/Nb:STO junction for HRS and LRS.

dependent transport for our Pt/Nb:STO junctions.

In summary, we investigated electrical properties of resistive-switching Pt/Nb-doped $SrTiO_3$ junctions. Comparative barrier height estimations using $I-V$ and photocurrent analyses showed that RS of the junctions varied the local fluctuation of the interface potential profiles, whereas the mean barrier height was not altered. RS also affected the dominant transport mechanism: thermal activation of carriers for HRS and tunneling for LRS.

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