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We presented a systemic investigation on the joint effect of gate bias and light illumination on a metallic $LaAlO_3/SrTiO_3$ interface in the temperature range from 15 K to 300 K. We showed that the photo excitation significantly enhanced the gating effect for the metallic two-dimensional electron gas. However, its effect is strongly temperature dependent; it is strong at low and high temperatures, and weak in the intermediate temperature range. There are evidences that the amplified gating effect stemmed from enhanced carrier depletion while the Hall mobility remains nearly unaffected. Acceleration of the gating process, together with a training effect marked by a strong dependence on gating history of the getting effect, is induced by repeating the electric cycling, indicating atomic reconfiguration due to oxygen migration and the memory of the migration paths. *Published by AIP Publishing.* https://doi.org/10.1063/1.5009790

The two-dimensional electron gas (2DEG) at the $LaAlO_3$/$SrTiO_3$ (LAO/STO) interface has attracted great interest in recent years because of its exotic properties,[1] such as 2D magnetism,[2] 2D superconductivity,[3] and enhanced Rashba spin-orbital coupling.[4] Moreover, these properties could be effectively tuned by the gating effect, leading to metal-to-insulator transition,[5] tunable superconductivity,[3,6] and dramatic variation of the magneto-transport behaviors.[7] Obviously, the gating effect has shown a great potential in unraveling the emergent phenomena at complex oxide interfaces.

For conventional semiconductor devices, the gate field takes effect through charging/discharging a capacitor formed by the gate electrode and the conducting interface. Here, only the process of charge transfer takes place. However, the gating effect is much more complex for complex oxide 2DEGs. In addition to charge transfer, many processes such as the migration of ionic defects, charge trapping/de-trapping, and ferroelectric instabilities could be involved, severely affecting the gating effect. As a consequence, the transport behaviors of the 2DEG have quite complex responses to gate field, yielding unforeseen phenomena. In the previous investigations,[8–10] it has been found that lattice deformation and a concomitant lattice polarization of STO can be induced by simultaneous application of gate field and light illumination, yielding a greatly enhanced gating effect on the semiconducting LAO/STO interface; the maximal increase in the sheet resistance caused by this combined gating effect is 200-fold larger than that produced by gate field alone. These works opened a wide space for the exploration of emergent phenomena.

However, there are several issues that remain to be addressed. The first one is what will happen to a metallic 2DEG under the combined stimuli of gate field and light illumination. As reported,[9] for semiconducting LAO/STO, the decrease in the photo excitation-induced resistance played a key role in enhancing the gating effect, especially at low temperatures. Different from semiconducting LAO/STO, the effect of photo excitation is obviously weak for a metallic 2DEG. The second issue is the effects associated with kinetic process of anion migration. As demonstrated, the enhanced gating effect originated from the lattice polarization due to the electro-migration of oxygen vacancies. In this scenario, distinct effects related to anion migration are expected while repeating electric cycling.

In this letter, we presented a systemic investigation on the joint effect of gate bias and light illumination on a metallic LAO/STO interface. We observed significantly enhanced gating effect by light illumination. The combined effect is yet strongly temperature dependent. We further found that the amplified gating effect stemmed from enhanced carrier depletion rather than the reduction of mobility. Moreover, acceleration of the gating process is induced by repeating the electric cycling, indicating a memory of the migration paths for oxygen vacancies after the removal of the applied field.

Samples were prepared by depositing a LAO layer (15 unit cells in thickness), using the technique of pulsed laser (248 nm) ablation, on TiO_2-terminated (001) STO substrates with the dimension of $2.5 \times 3 \times 0.5\,mm^3$. The deposition temperature was 750 °C, and the oxygen pressure was 10^{-5} mbar. The fluence of the laser pulses was 0.7 J/cm^2 and the repetition rate was 1 Hz. After deposition, the sample was naturally cooled to room temperature without changing oxygen pressure. The standard four-probe technique was employed for resistance measurements. Ultrasonic Al-wire bonding was

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adopted for electrical contacts. The applied current for the resistance measurements was 20 μA. Based on the formula $R_S \approx (L/W)R$, the sheet resistance (R_S) is deduced from the four-probe resistance (R), where L and W are the long and wide dimensions of the measured plane, respectively. For the gating effect study, a gate voltage (V_G) was applied to the back gate of STO while the LAO/STO interface was grounded, and the sheet resistance was recorded in the presence/absence of a light illumination. In all cases, the leakage current (<50 nA) was much lower than the applied current for resistive measurements (20 μA). The laser beam with the wavelength of 532 nm and a spot size of 1 mm in diameter was employed for the investigation of photo excitation. Capacitance was measured under an a.c. bias with the amplitude of 0.5 V and the frequency of 5 kHz. Hall effect was measured with a current of 10 μA in the field range from −1 T to 1 T, adopting the van der Pauw geometry.

Figure 1(a) presents the temperature dependence of the sheet resistance (R_S) of our 2DEG. A monotonic decrease in R_S with the decrease of temperature is observed. R_S is ~85.9 kΩ at 300 K and ~0.8 kΩ at 15 K. To determine the carrier density (n_S) of our 2DEG, we also measured the Hall resistance at several selected temperatures. As shown in Fig. 1(b), n_S varies from $\sim 9.2 \times 10^{12}\,\text{cm}^{-2}$ to $\sim 3.6 \times 10^{13}\,\text{cm}^{-2}$ as temperature increases from 15 K to 300 K. The deduced Hall mobility is 763.1 cm^2/V s at 15 K and 2.0 cm^2/V s at 300 K. These are typical features of a metallic 2DEG. It is different from the previously studied 2DEG for the joint effect of gate field and light illumination, which is semiconducting below 300 K.[8,9]

Figure 2(a) is a sketch of the measurement geometry. Figures 2(b)–2(d) show the temporal evolutions of the sheet resistances (R_S) recorded at three selected temperatures. Without light illumination, two distinct processes can be identified upon the application of a negative gate voltage, i.e., a sudden jump and a followed slow increase (or a fairly rapid decrease at lower temperatures) in sheet resistance. As revealed by previous investigations, the first process stems from the conventional capacitive effect that depletes the charge carriers of the 2DEG, and the following slow R_S growth is due to the electro-migration of oxygen vacancies, which results in lattice polarization. The resistance decay after the first jump at low temperatures is not observed before. It implies a slow charge transfer from the 2DEG to the in-gap states of LAO/STO when the energy band is inclined by negative gate biases. The resistance jump and the followed slow resistance decay after the removal of the positive bias is interesting. We ascribed the first process to the depletion of the 2DEG and the second process to a slow backward charge transfer from in-gap states to 2DEG due to the recovery from the declined band state caused by positive gate biases. These effects are strong at low temperatures and in a dark environment since the thermal and photo excitations of the in-gap states are not obvious in this case.

As expected, the incorporation of light illumination drives the first process into an enhanced sudden jump and, in the same time, accelerates the resistance growth of the second process. Reversing gate direction reverses the gating effect. Irrespective of gate direction, light illumination has a strong influence on gating effect. The maximal enhancement of the relative resistance change is as high as 1058%, occurring at 300 K. These phenomena are similar to those observed for the semiconducting LAO/STO interface.

To give a quantitative description to the gating effect, we define $\Delta R_S/R_S = [R_S(V_G) - R_S(0)]/R_S(0)$. Figure 2(e) shows the $\Delta R_S/R_S$ as a function of temperature. Without illumination, $\Delta R_S/R_S$ is ~9.8 at 15 K and monotonically decreases to ~0.2 as the temperature is increased to 200 K. This is understandable noting the monotonic decrease of the permittivity of STO as the temperature increases. From 200 K to 300 K, $\Delta R_S/R_S$ display a sizable upturn from 0.2 to 0.5. As will be discussed later, this implies the occurrence of lattice polarization. Here, ΔR_S is defined by the first resistance jump, which corresponds to the effect of charge depletion/accumulation caused by the gate field. In the presence of light illumination, remarkably, the variation of $\Delta R_S/R_S$ with temperature is no longer monotonic. $\Delta R_S/R_S$ takes the maximal value of ~22.4 at 15 K and rapidly decreases to ~4.5 when the temperature is increased from 15 K to 50 K. After that, it keeps nearly constant (~1.3) from 100 K to 200 K, and then increases to ~10.5 at 300 K. Here, ΔR_S is defined as the resistance difference after the gating of 200 s. This is in sharp contrast to semiconducting LAO/STO interface, for which the combined effect of the gate field and light illumination is nearly temperature independent.[9]

The above results demonstrated the acceleration and amplification of the gating effect by light illumination. As well known, gate biases modulate the sheet resistance by affecting the carrier density and mobility. In the previous reports on semiconducting LAO/STO, no direct evidence for the variation of carrier density and mobility was presented.[8,9] To reveal their respective effects, we simultaneously measured Hall resistance and sheet resistance as functions of gating time. We performed the experiments at room temperature where oxygen vacancies are more easily driven by gate field than at low temperatures. A small gate voltage of −10 V was applied to the back gate in the measurement process; a large V_G will cause a rapid variation in Hall resistance, making the measurement unreliable. Without the help of light illumination, as shown in Fig. 3, gate bias causes very small changes in carrier density and mobility after a gating time of 270 min, ~2.1% for the former and ~1.6% for the latter. With the use of light illumination, in contrast, it induces a dramatic variation in R_S, which has been amplified by a factor of 3.6. Accordingly, the carrier density

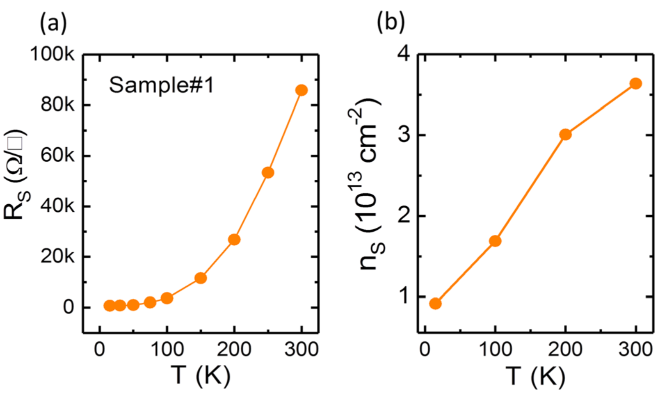

FIG. 1. (a) Sheet resistance as a function of temperature for one of our 2DEGs. A typical metallic behavior is shown. (b) Temperature dependence of carrier density, deduced from Hall resistance at selected temperatures.

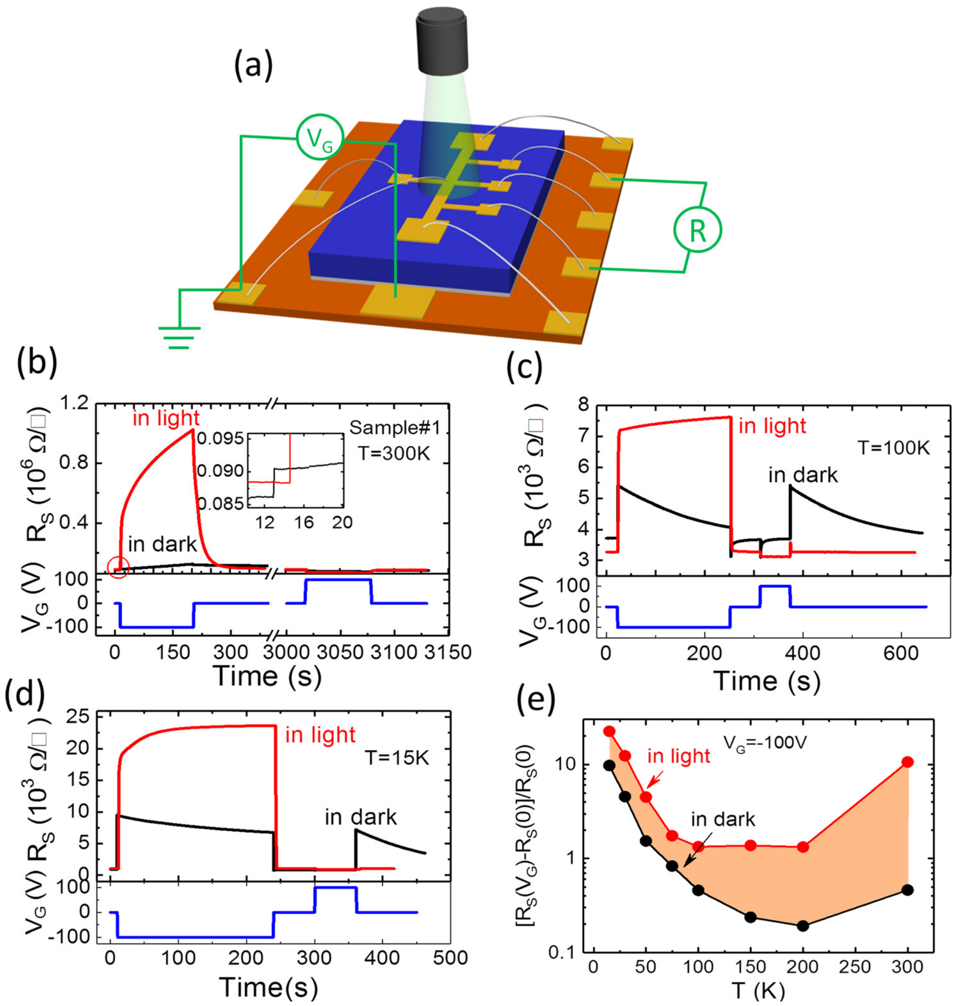

FIG. 2. Temporal evolution of sheet resistance corresponding to the application/removal of a gate voltage in the presence/absence of light illumination (light power = 10 mW, wavelength = 532 nm). (a) A sketch of the measurement geometry. (b)–(d) correspond to 300 K, 100 K, and 15 K, respectively. The inset plot in (b) is a close view of the encircled part of the R_S-T curve. (e) Temperature dependence of the relative resistance change. The carrier density of sample #1 is $\sim 3.6 \times 10^{13}$ cm^{-2} at 300 K.

varies from $\sim 3 \times 10^{13}$ cm^{-2} to $\sim 1 \times 10^{13}$ cm^{-2}, corresponding to a decrease of 67% [Fig. 3(b)]. This change is much greater than that expected by the capacitive effect ($\Delta n_S \sim 3.3 \times 10^{10}$ cm^{-2}). It is the direct evidence for the tuning of the charge carrier by the mechanism other than the normal capacitive effect. According to the works of Li *et al.*,[9,10] this additional mechanism is lattice polarization. Based on the formula of $\Delta n_S = P \cdot S$, the lattice-polarization-resulted electric polarization is calculated, and it is $P = 2\ \mu$C/cm^2 under a V_G of -10 V. Notably, this value is even greater than that obtained for the semiconducting LAO/STO under a V_G of -100 V ($\sim 1.6\ \mu$C/cm^2).[9] It means that the oxygen vacancies in the present sample are more mobile. This echoes our previous postulation that there are a plenty of in-gap states in our samples. As a comparison, it should be noted that without lattice polarization, the electric polarization of the STO is only 5.3×10^{-3} μC/cm^2 under a V_G of 10 V, i.e., it is the lattice polarization that dominates the gating effect.

In general, charge carriers farther from the interface will exhibit a higher mobility. When a negative electric bias is applied to the back gate, these charge carriers will be the first to be exhausted, leading to a decrease of the Hall mobility. Fascinatingly, our work suggests that there may be no preferential charge depletion for our 2DEG. This result indicates that the mobility of the charge carriers of our 2DEG may be more uniform, irrespective of their distance from the interface. This is different from the conclusion obtained by Bell *et al.*[6] who found a simultaneous variation in the carrier density and mobility. As shown in Fig. 3(c), the Hall mobility at 300 K undergoes a very small variation after being gated by a V_G of -10 V for 270 min, though $\Delta n_S/n_S$ is as high as $\sim 67\%$.

There are indications that the carrier mobility is also insensitive to the gate field at lower temperatures. Because of the limitation of experimental conditions, we cannot perform Hall resistance measurement at lower temperature in the simultaneous presence of gate biases and light illumination. As an alternative, we estimated Δn_S based on the capacitance-voltage relation [Fig. 3(d)] via the formula $\Delta n_S = 1/e\int_0^{V_G} CdV$, where C is the capacitance of the 2DEG-back gate capacitor, and e is the electron charge. As a representative, in Fig. 3(e), we show the deduced n_S under a V_G of -60 V (black symbols). Compared with its counterpart value at $V_G = 0$, n_S under $V_G = -60$ V is obviously lower. The maximal $\Delta n_S/n_S$ is $\sim 75\%$, occurring at 15 K. Ignoring the variation of Hall mobility, we obtained $\Delta R_S/R_S \approx -\Delta n_S/(n_S + \Delta n_S)$. Fascinatingly, this rough estimation well reproduced the gating effect in the temperature range from 15 K to 200 K [Fig. 3(f)], i.e., the change in n_S is the main origin of the gating effect. The discrepancy at 300 K can be attributed to lattice polarization which cannot be captured by capacitance.

Moreover, the gating effect is strongly history dependent: Repeated on/off cycling of gate bias accelerates the gating process. As shown in Fig. 4(a), R_S is initially $\sim 2.4 \times 10^4$ Ω at room temperature, and becomes 3.9×10^5 Ω after the first

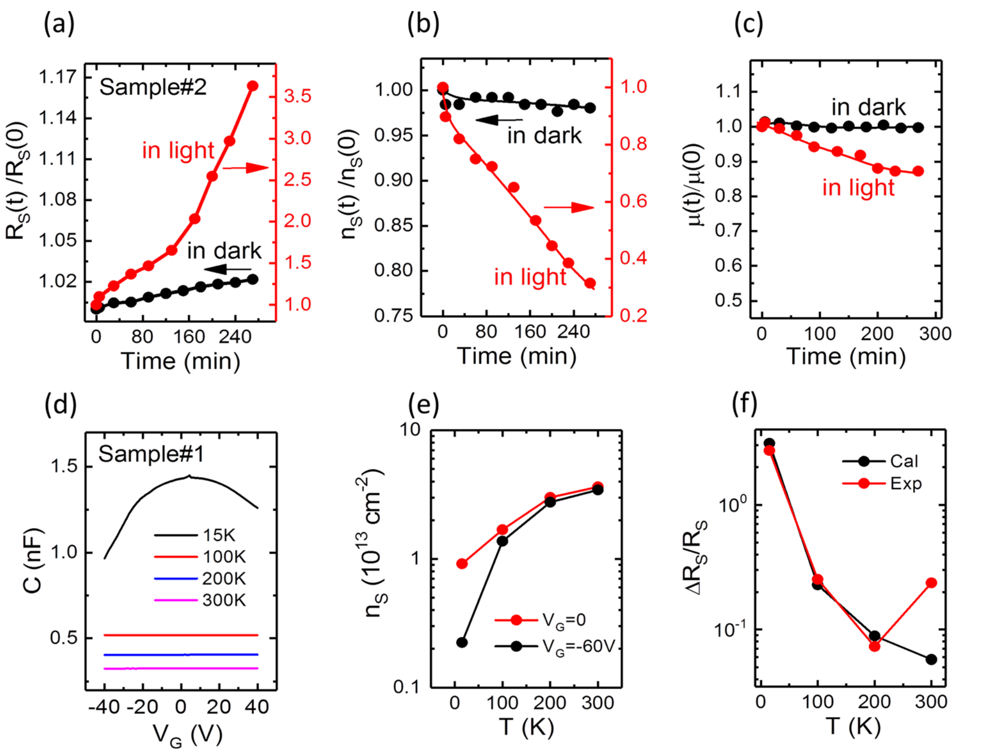

FIG. 3. Correspondence of normalized sheet resistance (a), carrier density (b), and Hall mobility (c) for sample #2 ($n_S \approx 3 \times 10^{13}\,\mathrm{cm}^{-2}$), measured as functions of gating time. Here, "in dark" and "in light" indicate the measurements without and with an additional laser illumination (10 mW, 532 nm), respectively. (d) Capacitance of the Ag/STO/LAO capacitor as a function of gate voltage for sample#1, measured at different temperatures. (e) Carrier densities corresponding to $V_G = 0$ and $V_G = -60\,\mathrm{V}$, respectively. Here, the $n_S(V_G = -60\,\mathrm{V})$ is deduced from the data in (d). (f) Resistance change caused by the application of $V_G = -60\,\mathrm{V}$. Here, red symbols represent the directly measured data and black symbols are results obtained based on $\Delta R_S/R_S \approx -\Delta n_S/(n_S + \Delta n_S)$ from the carrier densities in (e). At room temperature, the carrier density is $\sim 3.6 \times 10^{13}\,\mathrm{cm}^{-2}$ for sample #1 and $3.0 \times 10^{13}\,\mathrm{cm}^{-2}$ for sample #2.

200-second-gating and $3.4 \times 10^6\ \Omega$ after the fifth 200-second-gating, i.e., $\Delta R_S/R_S$ is amplified by a factor of 9.2. It implies that the migration of oxygen vacancies becomes easier and easier with the repetition of the electric cycles, i.e., the gating effect is strongly gating history. We call it the training effect. In Fig. 4(b), we show the $\Delta R_S/R_S$ under illumination as a function of the number of electric cycles at different temperatures. The training effect is obvious only at high temperatures.

To see what has happened to 2DEG, we traced the evolution of carrier density with electric cycling. The initial carrier density is $\sim 3.2 \times 10^{13}\,\mathrm{cm}^{-2}$, deduced from Hall resistance. Based on relation $\Delta R_S/(R_S + \Delta R_S) \approx -\Delta n_S/n_S$, the carrier density change after each gating can be roughly estimated. The first gating causes an Δn_S of $\sim 3 \times 10^{13}\,\mathrm{cm}^{-2}$, and the corresponding $\Delta R_S/R_S$ is ~ 13.6. The largest Δn_S appears after the fifth gating. It is $\sim 3.17 \times 10^{13}\,\mathrm{cm}^{-2}$, corresponding to a $\Delta R_S/R_S$ of ~ 121.3. Compared with the effect of the first gating, $\Delta R_S/R_S$ is enhanced by a factor of ~ 9 whereas the depleted carrier density $\Delta n_S/n_S$ is increased only ~ 0.05 [$\Delta n_S \sim 1.7 \times 10^{12}\,\mathrm{cm}^{-2}$, Fig. 5(a)]. By recording the discharging process after the removal of gate field, we obtained the variation of discharged charges (ΔQ) for different electrical cycles, and deduced the corresponding change in carrier density by $\Delta Q/eS$, where S is the area of the electrode. As shown in Fig. 5(a), the results deduced by discharging experiments confirmed the small variation of Δn_S. It means that the effect of electric cycle on carrier depletion is very weak though it is strong on the sheet resistance. This is an unexpected but understandable result. Since the charge carriers are nearly exhausted after the first gating, even a minor further loss in carrier density will cause a drastic increase in $\Delta R_S/R_S$. Therefore, a strong training effect may prefer to appear when the charge carriers are nearly completely exhausted or thermal energy is high enough to activate the migration of

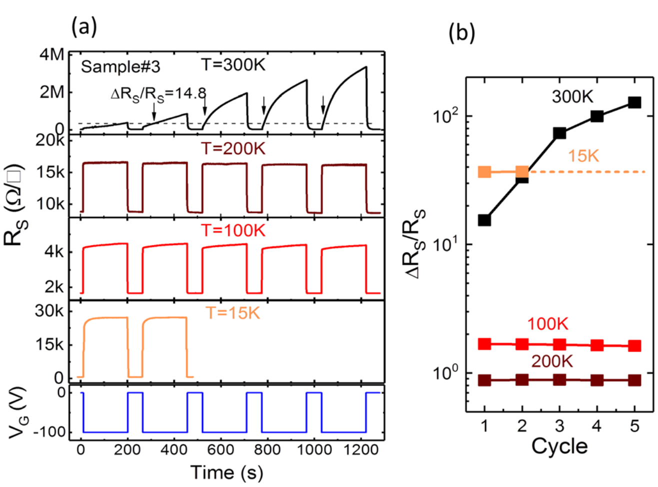

FIG. 4. (a) Evolution of the sheet resistance with time in response to the repeated on/off operations of a gate bias, recorded at four representative temperatures. All measurements were performed in the light of 10 mW (532 nm). (b) A summary of the resistance change at different temperatures, as a function of the number of electric cycles. The carrier density is $\sim 3.2 \times 10^{13}\,\mathrm{cm}^{-2}$ for sample #3 at 300 K.

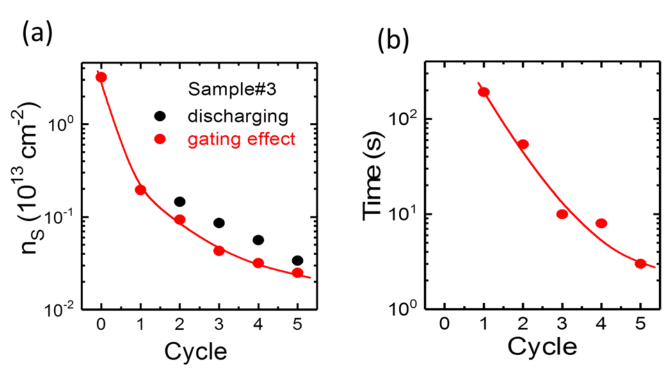

FIG. 5. (a) Carrier density as a function electric cycles (red symbols), deduced from the equation $\Delta n_S/n_S \approx -\Delta R_S/(R_S + \Delta R_S)$ from the data in Fig. 4(a) (red symbols) or from discharging experiment (black symbols). (b) Gating time required to get a $\Delta R_S/R_S$ of 14.8. Its decrease indicates the acceleration of the gating process.

oxygen vacancies. This explains why it is observed only at room temperature.

A further remarkable observation is the acceleration of the gating process by repeatedly electric cycling. As shown in Fig. 5(b), the required time to get a $\Delta R_S/R_S$ of 14.8 [dashed line in Fig. 4(a)] decreases from 192 s to 3 s from the first to the fifth gating. The gating process is speeded up by a factor of 64. Possibly, the electric field-driven anion migration causes considerable atomic re-configuration, leading to paths for oxygen vacancies. Although the virginal configuration could be recovered after the removal of gate field, these paths may be maintained to some extent. This result also implies that the lattice polarization process can be accelerated by repeating the electric cycles. It can also be realized at a relatively low gate field.

In summary, a systemic investigation on the joint effect of gate bias and light illumination on metallic $LaAlO_3$/$SrTiO_3$ interface is performed. A great enhancement of the gating effect by light illumination is observed. The joint effect is strongly temperature dependent. It is found that the amplified gating effect stemmed from enhanced carrier depletion while the Hall mobility remains nearly unaffected. The gating effect can be accelerated by repeated electric cycling, indicating atomic reconfiguration due to oxygen migration and the memory of the migration paths for oxygen vacancies. This work deepens the understanding of the photo excitation-enhanced gating effect at the LAO/STO interface. The principle proven here could also be extended to other complex oxide interfaces.

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