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Review of photoresponsive properties at $SrTiO_3$-based heterointerfaces*

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The two-dimensional electron gas at $SrTiO_3$-based heterointerfaces has received a great deal of attention in recent years owing to their potential for the exploration of emergent physics and the next generation of electronics. One of the most fascinating aspects in this system is that the light, as a powerful external perturbation, can modify its transport properties. Recent studies have reported that $SrTiO_3$-based heterointerfaces exhibit the persistent photoconductivity and can be tuned by the surface and interface engineering. These researches not only reveal the intrinsic physical mechanisms in the photoresponsive process, but also highlight the ability to be used as a tool for novel all-oxide optical devices. This review mainly contraposes the studies of photoresponse at $SrTiO_3$-based heterointerfaces.

Keywords: complex oxides, $LaAlO_3/SrTiO_3$ heterointerfaces, two-dimensional electron gas, persistent photoconductivity (PPC)

PACS: 78.67.Pt, 71.27.+a, 73.40.–c, 73.50.Pz **DOI:** 10.1088/1674-1056/27/11/117804

1. Introduction

In 2004, Ohmoto and Hwang reported an emergent two-dimensional electron gas (2DEG) at the interface between two well-known materials, $LaAlO_3$ (LAO) and $SrTiO_3$ (STO).[1] Later, some mechanisms, including polar discontinuity, oxygen vacancy, interface intermixing, are proposed to explain the presence of conduction at the interfaces.[2–10] The strong electronic correlations in 2DEG give rise to a variety of exciting properties, such as magnetism, gating superconductivity, enhanced Rashba spin–orbital coupling, tunable metal–insulator transition, and novel quantum Hall effect.[11–23]

In particular, external electrical fields, polar adsorbate, as well as photonic injection can manipulate the conductivity in this system by affecting the carrier at the interface.[24,25] As a powerful external perturbation, light also can modify the transport properties of complex oxides.[26–34] In fact, the sensitivity of LAO/STO to the light was discovered several years ago.[35] More recently, the investigation of transport properties of light-illuminated samples revealed its fundamental physics for potential applications, which has drawn a lot of attention.[36–43] For examples, Tebano *et al.* reported that the 2DEG at the LAO/STO heterointerface exhibited the giant persistent photoconductivity (PPC) in 2012.[44] In addition, an enhanced photoresponse by the surface modification using Pd nanoparticles was observed by Chan *et al.*[45] Particularly, the insulator–metal transition and the suppression of Kondo effect at the interface induced by light have been reported.[46–49] Apart from this, the obvious changes of photoresponsive properties of 2DEG at the LAO/STO interface by the substitution at Al sites have also been investigated.[50–54] In this review, we summarize the recent progress in the optical response of 2DEG at STO-based heterointerfaces.

2. Photoresponse in strontium titanate

STO single crystal is a band insulator with its bandgap ~ 3.25 eV at room temperature, having potential applications in oxide-based electronic devices.[55–57] When it is irradiated by the light with the photon energy higher than 3.25 eV, the photoinduced insulator–metal phase transition is observed. Under the illumination, STO shows a metallic conduction and the relative change in the resistance is above six orders at room temperature, as shown in Fig. 1(a).[58] Meanwhile, some defects at the surface can be produced by annealing STO at high temperature.[59–61] When exposed to sub-bandgap light at room temperature, the annealed samples exhibit a large PPC at room temperature as shown in Fig. 1(b), which is not observed in the as-received samples.[59] The free-electron concentration increases by over 2 orders of magnitude. Similarly, Ar^+-ion bombardment and laser irradiation on the insulating STO can also realize a metallic surface.[62,63] Oxygen deficiencies have been introduced to a depth of ~ 20 nm from the crystal surface. These deficiencies generate conduction carriers and stabilize a hole level in a self-trapped state. Kan *et al.* reported on blue-light emission at room temperature from irradiated samples.[57] They proposed that the doped conduction electrons and the in-gap state produce a radiative process that

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results in the blue-light emission. In addition, these irradiated samples display the ultraviolet light sensitive photoconductivity. This behavior maps well with the temperature dependence of the STO dielectric function. The origin of PPC is attributed to the excitation of an electron from a titanium vacancy defect into the conduction band with a very low recapture rate.

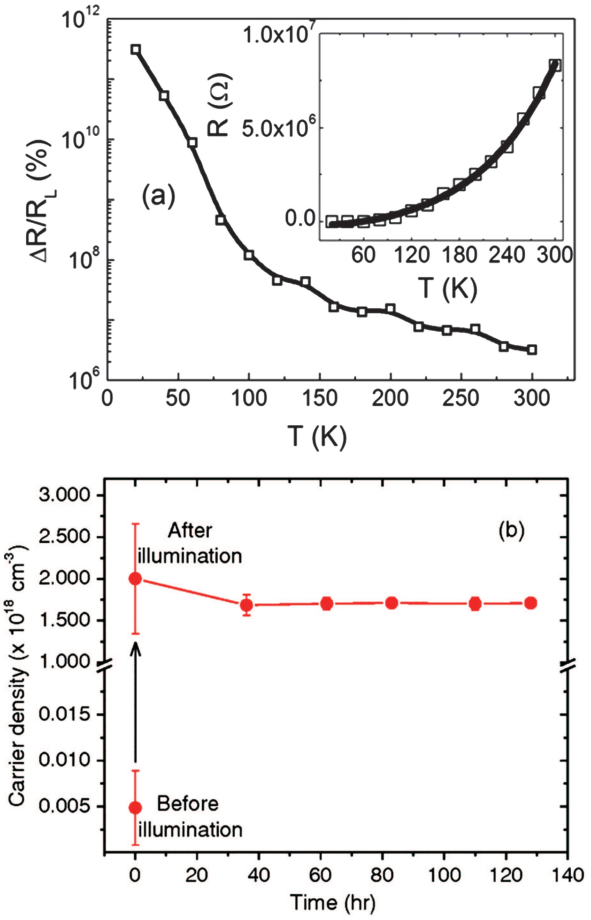


Fig. 1. (color online) (a) The photoinduced relative change in the resistance as a function of temperature. The inset shows that resistance vs. temperature curve of STO single crystals under the light irradiation. The solid line is the best fit to the data.[58] (b) Carrier density of the annealed sample before and after illumination. After the illumination, the sample was kept in the dark.[59]

3. Photoresponse at LAO/STO interfaces

3.1. PPC effect

The LAO/STO interfaces exhibit a giant PPC effect at room temperature in an ambient environment.[44] The conductance is increased by about 5 orders of magnitude. Photoinduced effects are observed under the UV (395 nm) and visible light illuminations, as shown in Fig. 2. The value at the LAO/STO interface is at least 2 orders of magnitude larger than the largest reported value for rough Si membranes in the literature.[64] The giant PPC effect at the LAO/STO interface can be explained qualitatively by the same mechanism active in the case of semiconducting interfaces. The energy associated with the wavelength of the radiation source is just below the band gap of STO. Hence, it cannot excite hole/electron pairs. On the other hand, residual oxygen vacancies behave as electron traps for the electrons. These traps are located inside the band gap of STO.[10,65] Because of the positively charged layer at the interface of a TiO_2-terminated STO substrate and the negative electronic space charge region responsible for the 2DEG, there exists the macroscopic potential barrier. Once a free electron has been generated by the absorption of a photon, the macroscopic potential barrier spatially separates the photoexcited electron from the parent donor defects. Thus, it is brought to close to the interface where all traps are already neutralized, giving rise to the PPC phenomenon.[66]

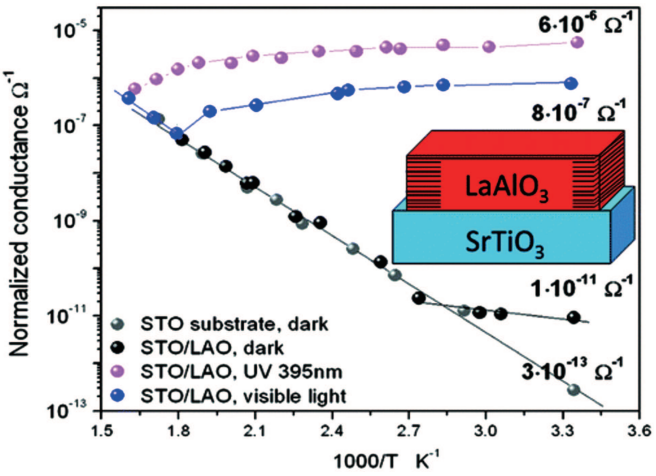


Fig. 2. (color online) LAO/STO normalized conductance as a function of the reciprocal of temperature, after the dark annealing at 573 K, still shielding the sample from any light (black dots), and illuminating the sample by a UV lamp of 395 nm (pink dots) or by visible light (blue dots). Also the STO substrate conductance (gray dots) is reported for comparison.[44]

Guduru *et al.* have illuminated LAO/STO interfaces with the light in the energy range between 1.44 eV and 3.65 eV, as shown in Fig. 3.[67] The influence of photons with different energies is clearly seen as a series of steps in the resistance of the sample. They attributed the small resistance change induced by illumination with energies smaller than the STO energy gap to the finite absorption of incident illumination by the in-gap states present inside the STO band gap. Differently, the low-temperature resistance even decreases by more than 50% after illumination with 3.65 eV. They explained the observations by the optical excitation of an additional high mobility electron channel, which is spatially separated from the photoexcited holes. The Hall resistance data after the illumination shows one low mobility (μ_1, 3 $cm^2/V{\cdot}s$) band with a high carrier density (n_1, $\sim 10^{14}$ cm^{-2}) corresponding to the original conduction band presenting before illumination, and one persistently photoexcited high mobility (μ_2, 1200 $cm^2/V{\cdot}s$) band with a low carrier density (n_2, $\sim 10^{10}$ cm^{-2}). For the Hall fitting, they ignored the possible change in the initial low-mobility carrier after illumination and therefore it cannot fully reveal the details. But it clearly points out the existence of an optically excited high mobility channel.

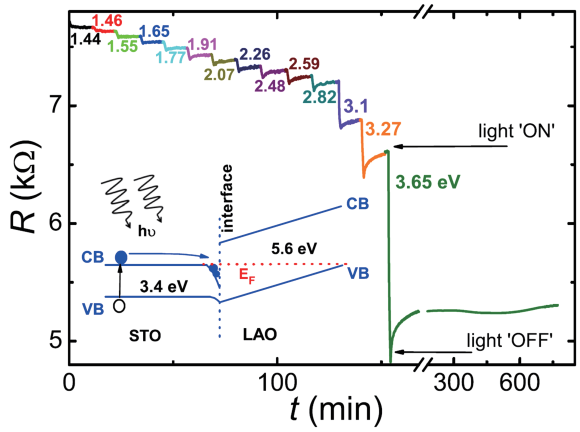


Fig. 3. (color online) Resistance as a function of time during the illumination with the photons energy from 1.44 eV to 3.65 eV at 4.2 K. Each change in the photon energy results in a pronounced step in the sample resistance; the photon energies, in eV, are shown beside each of the steps. Note the break on the time axis showing the persistence of the resistance change. The inset shows a schematic band diagram (CB: conduction band, VB: valence band, and E_F: Fermi-level) for a LAO/STO heterostructure under the illumination and presuming an internal potential build up in the LAO.[67]

Subsequently, we reported the photoresponsive relaxation characteristics in detail, as shown in Fig. 4.[49] The resistances quickly decrease to the minimum values when the light is on, and then show a decay to a steady value when the light is off. The persistent and transient photoinduced effects are simultaneously observed. Moreover, we analyzed the PPC effect from the band bending. Electrons are directly promoted from the VB maximum to the CB minimum under the irradiation of 365 nm light. The photoexcited electrons drift to the channel region, increasing the 2DEG density and then contributing to the observed decrease in the resistance. When the light illumination is off, the photoexcited electrons will be recombinated with holes at the VBM in STO and be recaptured slowly by the subbands.

Until now, most of previous attempts are focused on the change of electrical conductance.[38,39] In order to reveal the intrinsic dynamics and mechanisms of the photogenerated carries, the time dependent photo-Hall measurements have been performed, which are used to obtain the time dependence of the density and mobility of the photogenerated carriers.[68] After the illumination, a marginal decrease in n_2 and an increase in μ_2 are observed. The n_2 plummets from 7.8×10^{13} cm^{-2} to 5.0×10^{13} cm^{-2} and the μ_2 rises from 17.2 cm^2/V·s to 63.7 cm^2/V·s at 10 K (Fig. 5). It is found that the density and mobility after the light illumination obey a stretched exponential expression, further indicating that the variation of mobility caused by the electron–electron scattering plays an important role in the recovery process in addition to the reduction of carrier density.[69] Meanwhile, a non-linear Hall resistance at the LAO/STO interface under the illumination of a 360 nm laser at low temperature is observed. Additionally, the electric gating effect can tune the recovery process after light illumination and induce a disappearance of the non-linear Hall resistance.

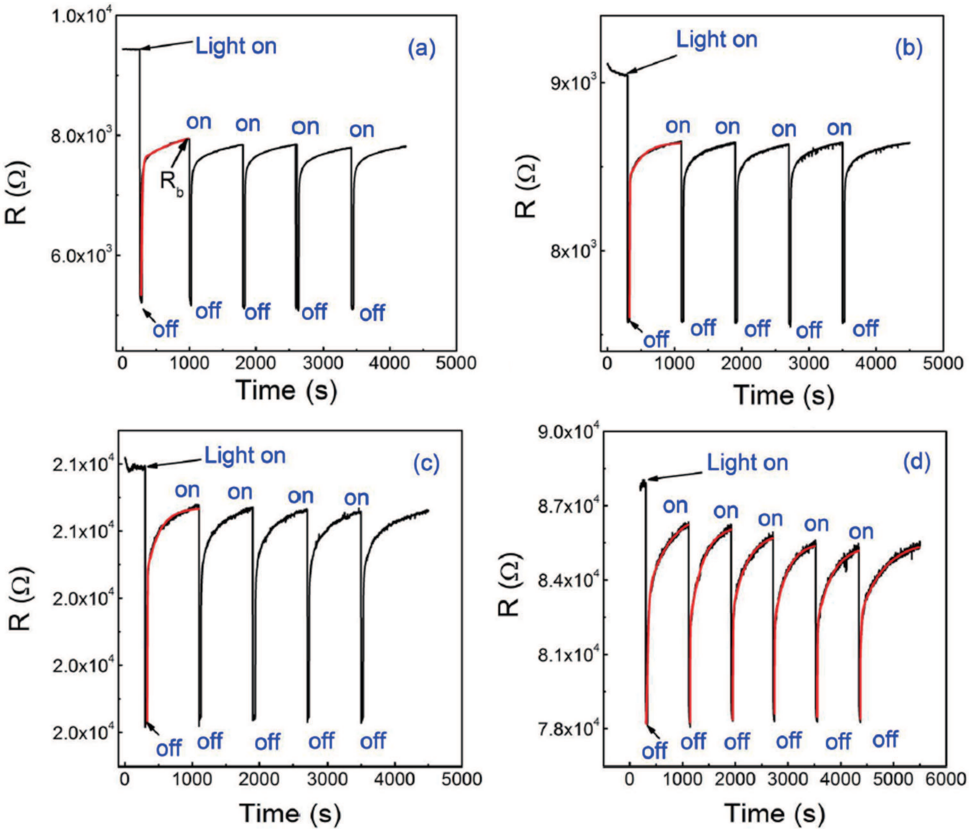


Fig. 4. (color online) Time dependence of the resistance at different temperatures: (a) 20 K, (b) 80 K, (c) 160 K, (d) 300 K. The red solid lines are the fitting curves.[49]

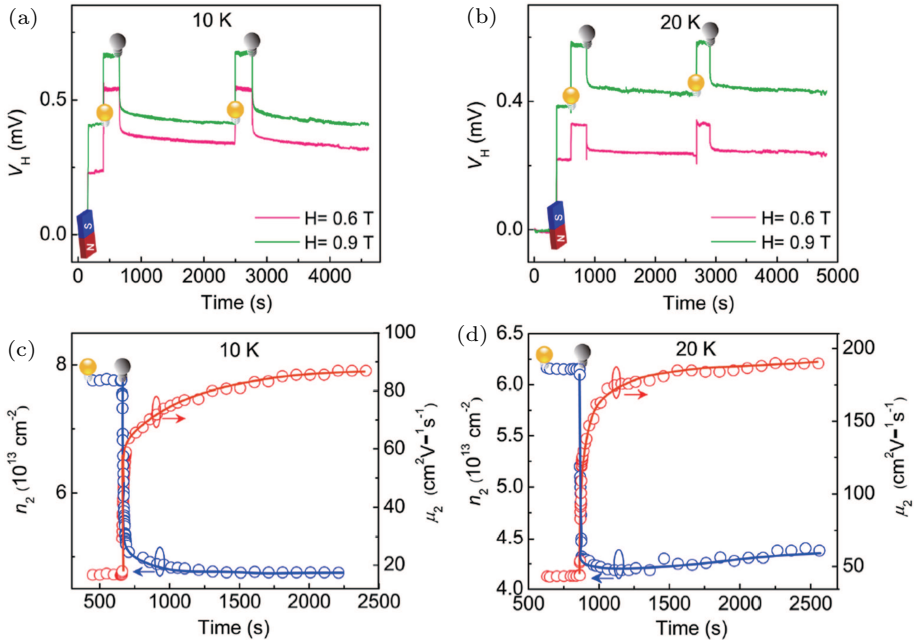


Fig. 5. (color online) (a) and (b) Time dependence of calculated Hall voltage V_H upon the application of $H = 0.9$ T and 0.6 T at 10 K and 20 K, respectively. (c) and (d) Time evolution of photogenerated carrier density (n_2) and Hall mobility (μ_2) at 10 K and 20 K, respectively.[68]

3.2. Modulation of PPC effect

3.2.1. Growth parameter

It is well known that the conduction of LAO/STO is extremely sensitive to the deposition condition, in particular to the oxygen background pressure and the heating temperature.[70] So these parameters also play a crucial role in the photoresponsive properties of oxide films and heterointerfaces.[71,72] Liu *et al.* have demonstrated the photoconductivity at the LAO/STO interfaces deposited at different oxygen pressures, as shown in Fig. 6.[73] The photoconductivity decreases rapidly with increasing oxygen vacancies in the heterointerfaces. In addition, the sample prepared at higher oxygen pressure exhibits the slower photorelaxation process due to the reduction of oxygen vacancies related in-gap states.

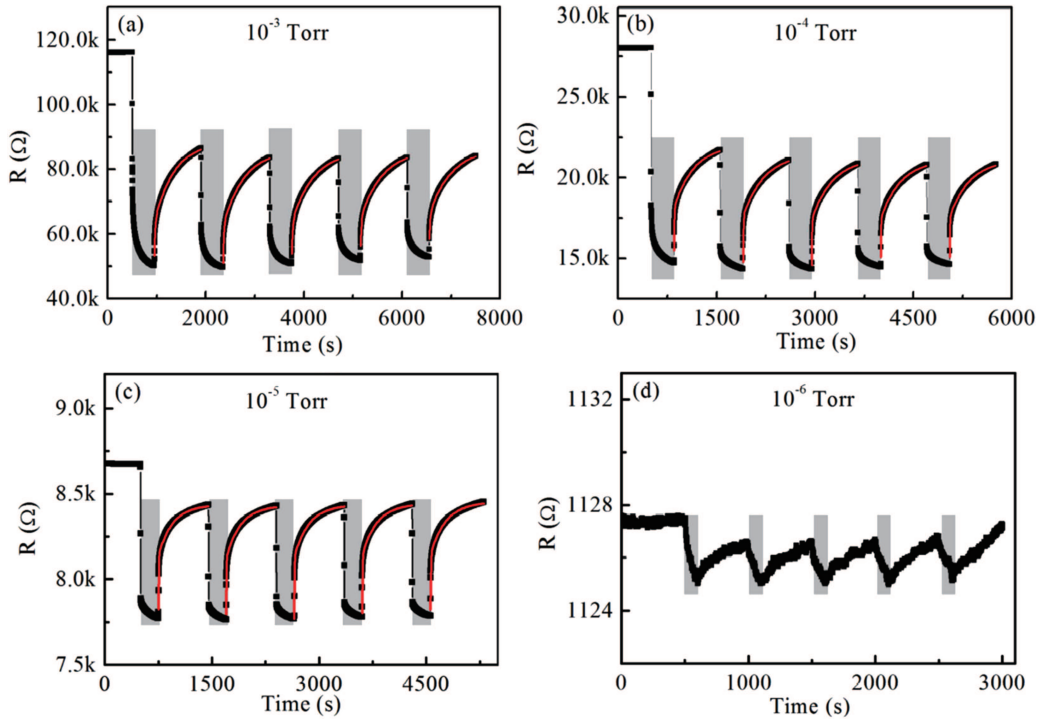


Fig. 6. (color online) Time dependence of the resistance for LAO/STO heterointerfaces fabricated at (a)–(d) oxygen pressures from 2×10^{-3} to 2×10^{-6} Torr. The red solid lines are the fitted curves and the grey areas show the time for light-on.[73]

In addition, Rastogi *et al.* have reported the photoconductivity in ultrathin films of LAO grown at several deposition temperatures.[38] The photoresponse clearly drops with lowering the deposition temperature, as observed in Fig. 7, suggesting that the ionization of oxygen defects by light is responsible for the enhanced conductivity. The number density of such defects will become less at a lower growth temperature.

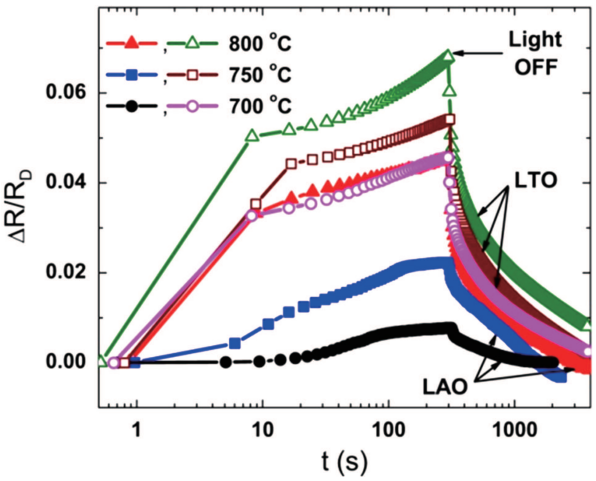


Fig. 7. (color online) Variation of the relative change in the resistance ($\Delta R/R_D$) with the time for 8 nm thick $LaTiO_3$ (LTO) (open symbols) and LAO (filled symbols) deposited at different temperatures on (100) STO substrates. These measurements were done at room temperature.[38]

Besides the parameters mentioned above, there are also reports that the thickness and strain of LAO film have significantly influences on the transport properties at LAO/STO interfaces.[74,75] Moreover, the photoresponse of LAO/STO interfaces with critical thickness (3 u.c.) has drawn a lot of interests and already been investigated.[47] Recently, we are studying the effect of the film thickness and strain on the photoresponsive properties and some meaningful results have been obtained.

3.2.2. Surface treatment

Since the 2DEG is confined to a ultrathin layer underneath LAO, a slight change on the LAO surface can significantly modulate the performance of 2DEG, such as via polar molecule adsorption or coating layer.[76–79] Brown *et al.* achieved a fully reversible conductance change at LAO/STO interfaces, regulated by LAO surface protonation.[80] As shown in Fig. 8, Pd nanoparticles with a size of around 2 nm are deposited on the top of LAO (5 u.c.) surface.[45] The change in the photocurrent of bare sample at 380 nm is only 18%. But for the Pd nanoparticles-coated LAO/STO sample, the change is about 750%, producing a giant photoconductivity. This giant optical switching behavior has been explained by the Pd nanoparticle's catalytic effect and surface/interface charge coupling. Pd nanoparticles attract electrons from the interface. When the UV light irradiates the surface, more electrons are induced at the 2DEG interface due to the generation of the photonexcited electrons and holes in the STO substrate. Pd nanoparticles absorb holes by the tunneling process at the surface, which is responsible for the enhanced photoconductivity.

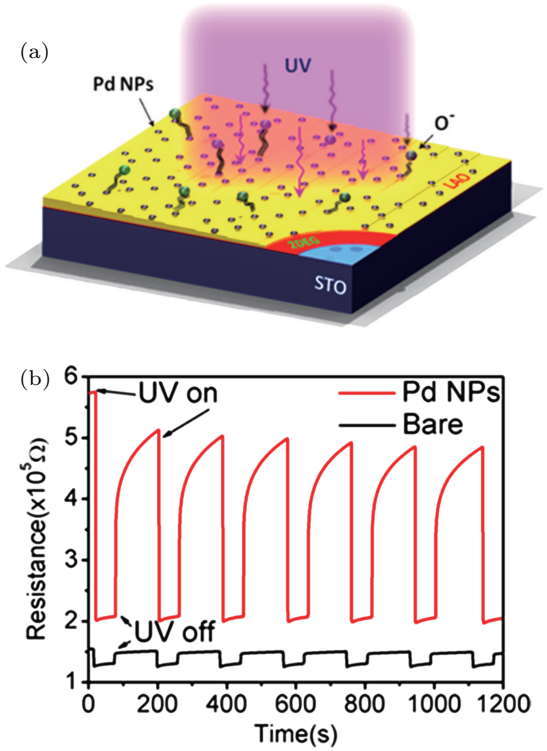


Fig. 8. (color online) (a) Schematic diagram of the Pd nanoparticle surface modulated LAO/STO with UV light irradiation. (b) Photoresponsive characteristics of the Pd nanoparticle-coated LAO/STO (red) and LAO/STO (black) heterointerfaces, showing the reversible switching behavior under periodic illumination of a 365 nm UV light with an incident power density of 10 mW/cm^2.[45]

3.2.3. Doping effect

Substitution or dopant is an effective method to tune the properties of LAO/STO interface.[51–53] In particular, Kumar *et al.* have reported a metal-to-insulator transition and a gradual suppression of 2DEG at the LAO/STO interface by the substitution of Cr at Al sites, producing a distinct change of the photoresistive properties in the $LA_{0.6}Cr_{0.4}O$/STO heterointerfaces.[54] We have modified the Kondo behavior by doping Ni and other magnetic elements at Al sites.[81,82] Under a 360 nm light irradiation, the interfaces exhibit a PPC effect and a suppressed Kondo effect at low temperature due to the increased mobility, as shown in Fig. 9. And the substitution causes a giant increase in the light-induced resistance change.

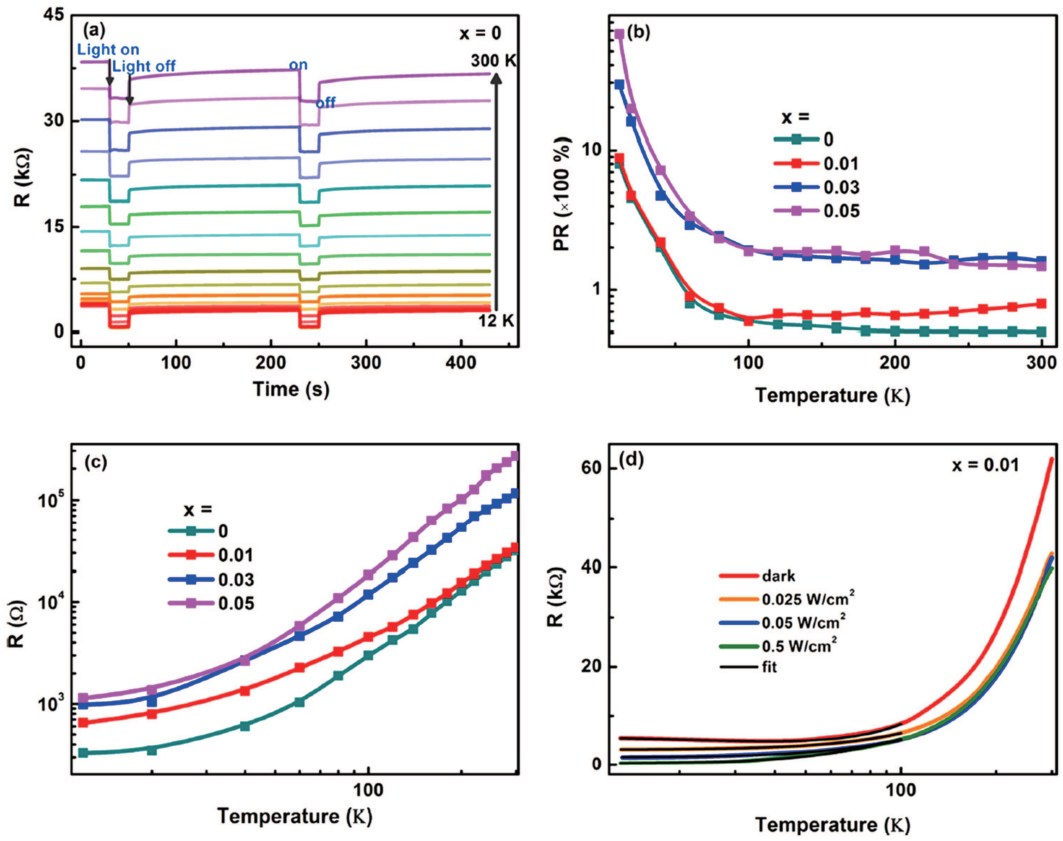


Fig. 9. (color online) (a) Time evolution of the resistance at Ni doped LAO/STO ($x = 0$) heterointerfaces under the irradiation of light with a power density of 0.5 W/cm^2 at different temperatures. (b)–(d) Temperature dependence of PR values, resistance under the irradiation (0.5 W/cm^2), and resistance under the irradiation with different power densities, respectively.[81]

3.2.4. Interface engineering

A systematic metal-to-insulator transition in the LAO/STO system has been observed by δ doping at the interface.[83–85] Rastogi *et al.* have reported the effect of δ doping at the LAO/STO interface with $LaMnO_3$ monolayers on the photoconducting state (Fig. 10).[86] Besides the significant modification in the electrical transport which drives a metal-to-insulator transition with increasing $LaMnO_3$ submonolayer thickness, an enhancement in the photoresponse and relaxation time constant is also observed. The possible scenarios for the photoconductivity are the defect clusters, random potential fluctuations, and large lattice relaxation models, along with the role of structural phase transition in STO.[87,88] For the pure LAO/STO, the photoconductivity originates from the interband transitions between Ti-derived 3d bands, which are e_g in character, and O 2p–Ti t_{2g} hybridized bands. The band structure is changed significantly when fractional layers of $LaMnO_3$ are introduced. Here, the Mn e_g bands (≈ 1.5 eV above the Fermi energy) within the photoconducting gap lead to a reduction in the photoexcitation energy and a gain in the overall photoconductivity. Besides, Liu *et al.* reported that the $SrRuO_3$ inserting at the LAO/STO interface induces a giant photoresponse.[89] All these researches provide a simple method to design 2DEG structures for future optoelectronic devices.

3.2.5. Electronstic gating effect

Electrostatic gating field is widely used a stimulus for semiconductor devices.[90–92] Via the capacitive effect, a gating field modifies the carrier density of devices, whereas the illumination generates extra carriers by exciting trapped electrons. Lei *et al.* have reported an unusual illumination-enhanced gating effect at LAO/STO interfaces, which has been the focus of emergent phenomena (Fig. 11).[20] Later, Yang *et al.* conducted the experiments at different temperatures to perfect the results.[93] It is found that the light illumination decreases, rather than increases, the carrier density of gas when the interface is negatively gated through the STO layer. The density drop is 20 times as large as that caused by the conventional capacitive effect. Further, this effect stems from an illumination-accelerated interface polarization, which is an originally extremely slow process. This unusual effect provides a promising control of correlated oxide electronics, in which a larger gating capacity is demanding due to their intrinsic larger carrier density. In addition, it has also been reported that optical gating can highly tune the spin–orbit coupling at the STO-based interfaces, which paves a way for designing optically controlled spintronic devices.[23]

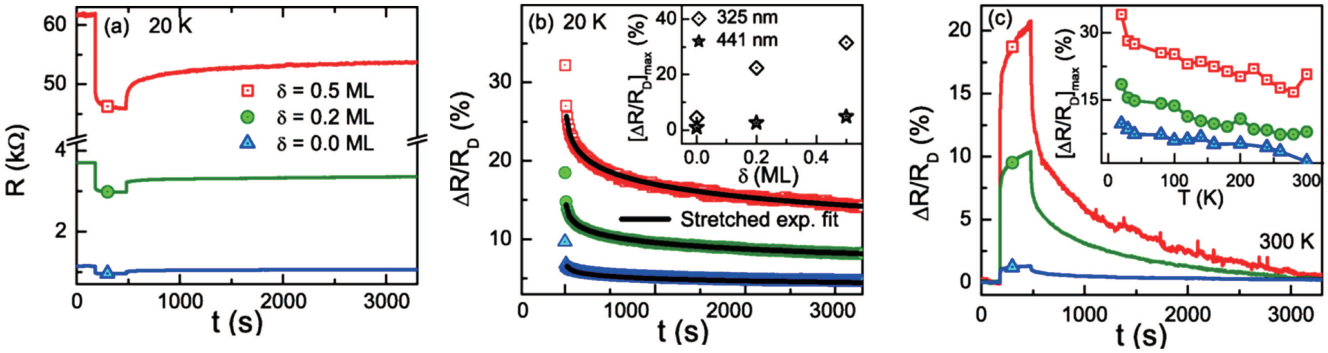


Fig. 10. (color online) (a) The change in the channel resistance at 20 K as a function of δ-layer thickness. Panels (b) and (c) respectively show the relaxation of normalized resistance for different δ doping at 20 K and 300 K after switching off the illumination from a halogen lamp. The recovery dynamics follow a stretched exponential behavior, which is represented as solid lines in (b). The inset of (b) shows the relative change in the resistance at 300 K upon irradiating the samples with 325 nm and 441 nm lines of a He–Cd laser. A comparison of the photoresponse as a function of temperature for different samples is made in the inset of (c). The 0.5 monolayer $LaMnO_3$ shows a three-fold increase in the photoresponse by comparison with the $\delta = 0$ sample.[86]

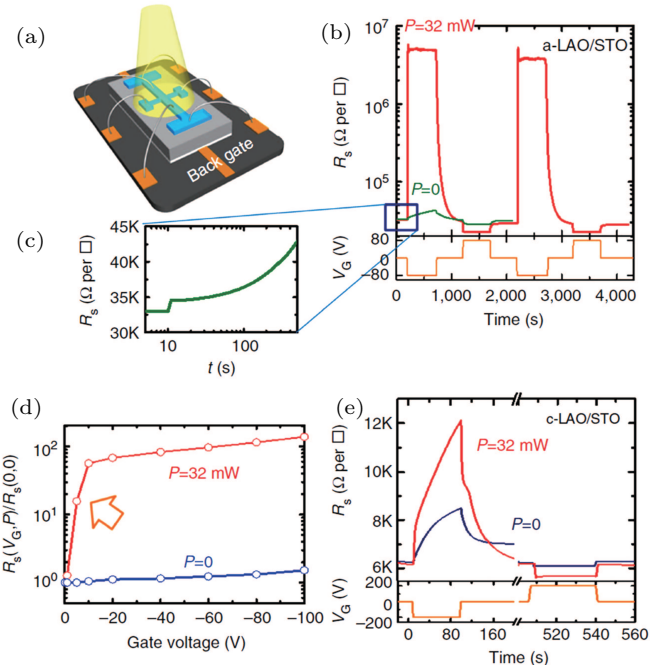


Fig. 11. (color online) Resistive responses to electrical and optical stimuli at the LAO/STO interface. (a) A sketch of the experimental set-up. (b) Sheet resistance of a-LAO/STO, recorded in the presence/absence of a light of $P = 32$ mW ($\lambda = 532$ nm) while V_G switches among -80 V, 0 V, and $+80$ V. (c) Enlarged view of the two-step feature of R_S without light illumination. (d) Gate dependence of normalized sheet resistance, $R_S(V_G,P)/R_S(0,0)$, recorded at the time of 300 s after the application of V_G. The arrow marks the R_S corresponding to $V_G = -5$ V. (e) Sheet resistance of c-LAO/STO, recorded in the presence/absence of a light of $P = 32$ mW ($\lambda = 532$ nm) as V_G switches among -200 V, 0 V, and $+200$ V. All measurements were conducted at room temperature.[20]

4. Conclusion and perspectives

We summarize the current researches on photoresponsive effect at STO-based 2DEG. The tunable photoresponsive properties at LAO/STO interfaces provide the potential and possibility for the application of photoelectric devices of all-oxides. Compared with traditional semiconductors, complex oxides have evident advantages because of their various functionalities and couplings, such as ferroelectricity, ferromagnetism, even superconductivity. Furthermore, there remain many open questions. For examples, the origin of 2DEG is still controversial. Acting as a method, the photoresponsive behavior might contribute to reveal the intrinsic mechanism by investigating the PPC effect or transient photoconductivity. In addition, the spin polarity is a desired nature, especially in 2DEG system, which is beneficial to the spintronics of all-oxide devices. To date, the spin direction of electrons in 2DEG system is disordered. Meanwhile, it can be expected that the photoresponse combined with the ferromagnetism in 2DEG system will be used in novel devices of magnetic-optical-electric response. So far, the 2DEG has been observed in many oxides, such as $LaGaO_3$, $LaTiO_3$, $GdTiO_3$, Al_2O_3, and $KTaO_3$.[94–98] Other 2DEG systems with higher mobility need to be explored further. Thus, the differences in the photoresponsive properties among these 2DEG systems require more researches. Ultimately, more work will need to be done in realizing the real application of oxides optoelectronics based on 2DEG systems.

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