Visible-light-enhanced gating effect at the LaAlO$_3$/SrTiO$_3$ interface

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Electrostatic gating field and light illumination are two widely used stimuli for semiconductor devices. Via capacitive effect, a gate field modifies the carrier density of the devices, while illumination generates extra carriers by exciting trapped electrons. Here we report an unusual illumination-enhanced gating effect in a two-dimensional electron gas at the LaAlO$_3$/SrTiO$_3$ interface, which has been the focus of emergent phenomena exploration. We find that light illumination decreases, rather than increases, the carrier density of the gas when the interface is negatively gated through the SrTiO$_3$ layer, and the density drop can be 20 times as large as that caused by the conventional capacitive effect. This effect is further found to stem from an illumination-accelerated interface polarization, an originally extremely slow process. This unusual effect provides a promising controlling of the correlated oxide electronics in which a much larger gating capacity is demanding due to their intrinsic larger carrier density.
The two-dimensional electron gas (2DEG) at the heterointerfaces between complex oxides has received attention in recent years because of its implementation for novel physics and prospective applications. The 2DEG confined to the LaAlO$_3$/SrTiO$_3$ (LAO/STO) interfaces is a representative system that has been extensively studied, and exotic properties including two-dimensional superconductivity, magnetism, enhanced Rashba spin–orbital coupling and strong electrical field effect have been observed. Among these, the field effect is particularly interesting. As already demonstrated, the transport behaviour can be tuned by a gate field across STO or LAO, undergoing a metal-to-insulator transition or a tunable superconducting transition. On the other hand, a dramatic modification of the interfacial conductivity can also be gained by depositing polar molecules or charges above the LAO layer. Obviously, gating effect has shown its potential in unravelling the emergent phenomena at complex oxide interfaces.

However, the electrical field effect for the complex oxide 2DEG is much more complicated than for the conventional semiconductor devices. In addition to electrons, there are many other factors such as ionic defects, trapped charges or ferroelectric instabilities in the system that can be severely affected by the applied electric field. As a consequence, significant hysteresis of the interfacial conductivity can occur when cycling electrical bias through the STO crystal or scanning a biased tip across the LAO layer, the latter leads to conducting nanowires persisting for days. A few recent reports even show that the field effect actually exhibits two steps: a fast process is followed by an extremely slow process that usually lasts for thousands of seconds but owns a tuning ability comparable to or even stronger than the fast one. While the slow process yields additional freedom in controlling the physical properties of the 2DEG, its slow nature makes it hard to be exploited in any practical devices but causes an adverse influence to the reproducibility.

In this work, we report on a remarkable effect produced by combined electrical and optical stimuli for the 2DEGs at both amorphous and crystalline LAO/STO heterointerfaces (a-LAOSTO and c-LAOSTO, respectively). We found that an illumination of visible light drives the slow field-induced resistance growth into a great jump far beyond the scope of normal field effect, markedly enhancing the ability of the gate field to modulate charge carriers. The present work clearly demonstrates the mutual reinforcement of the effects of electrical gating and light illuminating on complex oxide interfaces.

**Results**

**Illumination-accelerated gating effect.** Details for sample fabrication and resistive measurements are described in the Methods. Figure 1 shows the typical resistive responses of our devices to electrical and optical stimuli. As schemed in Fig. 1a, a gate voltage,
$V_G$, between $-100$ V and 100 V was applied to the back gate of STO while the a-LAO/STO interface was grounded and the sheet resistance, $R_s$, was recorded in the presence/absence of a light illumination. In all cases, the leakage current ($<7$ nA) was much lower than the in-plane current applied for resistive measurements, 1 µA (Supplementary Fig. 1). As shown in Fig. 1b,c, without illumination, the application of $V_G = -80$ V yields two distinct processes marked respectively by a slight jump and a followed steady increase of $R_s$. The first minor jump is the normal gating effect, stemming from the field-induced charge density change in the backgate–interface capacitor. The latter process is extremely slow, lasting for $>2000$ s without saturation and produces an $R_s$ increase much larger than the first jump. This process can be well described by the Curie–von Schweidler law $R_s \propto (1 - n_0)^2$, which implies a wide distribution of the energy barriers that impede the carrier depletion (see Supplementary Fig. 2).

Remarkably, such a field effect is significantly modified by light illumination. Aided by a light of $32$ mW ($\lambda = 532$ nm), as shown by the red curve in Fig. 1b, gate field drives $R_s$ into a sudden jump to a steady state of 200-fold resistance, that is, the slow process has been markedly accelerated by light illumination, and this change is reversible for the repeated on–off operations of the gate field. As summarized in Fig. 1d, a light of 32 mW pushes the $R_s$($V_G = -100$, $V_P$)/$R_s$(0,0) ratio from $\approx 1.2$ to $\approx 202$, amplifying the gating effect by a factor of 170-fold. Moreover, even a $V_G$ as low as $-5$ V can cause $R_s$ growth (marked by an arrow). This bias is only one-tenth of that usually required to get comparable effect using a backgate without light.

The gating effect of positive $V_G$ was also enhanced by illumination but it is relatively weak (see Fig. 1b and Supplementary Figs 3 and 4). Similar illumination-enhanced gating effect is also observed in c-LAO/STO (Fig. 1f), suggesting that it is a quite universal phenomenon, independent of the characteristics of the electronic transport of the interface (it is semiconducting for a-LAO/STO and metallic for c-LAO/STO; refer to Supplementary Fig. 5) and the crystal structure of the LAO overlayer (crystalline or amorphous).

**Carrier density tuning beyond capacitive effect.** To gain a further understanding of this illumination effect, we examined the sheet carrier density, $n_s$, by Hall measurement. From the linear $R_{xy}$–$H$ relation in Fig. 2a, the initial $n_s$ can be deduced, and it is $\approx 7 \times 10^{12}$ cm$^{-2}$, where $R_{xy}$ is the Hall resistance. There are no detectable changes in the $R_{xy}$–$H$ dependence measured immediately after the application of a $|V_G| = 100$ V, indicating that the change in carrier density is tiny. It is consistent with the result deduced from the capacitance data in Fig. 2b. Here, $\Delta n_s = 1/\epsilon_0 \int_{-100V}^{0} C_{a-LAO/STO} dV \approx 3 \times 10^{11}$ cm$^{-2}$ for $P = 0$, which is only $\approx 4\%$ of the initial $n_s$, where $C_{a-LAO/STO}$ is the capacitance of the backgate–interface capacitor, $\epsilon$ is the electron charge and $S \approx 5$ mm$^2$ is the interface area. In contrast, in a light of $P = 6$ mW (the highest intensity available for our Hall-effect measurement system), a $V_G = -100$ V reduces the $n_s$ from $\approx 7.0 \times 10^{12}$ to $\approx 1.3 \times 10^{12}$ cm$^{-2}$ (Fig. 2b) and the mobility from $\approx 25.8$ to $\approx 1.2$ cm$^2$ V$^{-1}$ s$^{-1}$ (deduced from the data in Fig. 2b). This extraordinarily large $\Delta n_s$ is confirmed by the sudden $C_{a-LAO/STO}$

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**Figure 2 | Hall effect and capacitance measurements.** (a) Hall resistance, $R_{xy}$, of a-LAO/STO measured with an in-plane current of 10 µA under different gating/illuminating conditions. Without light illumination the data for $V_G = -100$ V cannot be distinguished from those for $V_G = 0$, and therefore are not shown here. (b) Carrier density and sheet resistance as functions of light power, acquired under a fixed $V_G$ of $-100$ V. Solid lines are guides for the eye. Dashed line is the extrapolated $n_s$–$P$ relation. (c) Capacitance, $C_{a-LAO/STO}$ of a-LAO/STO as a function of gate voltage, measured under the a.c. amplitude of 0.5 V and frequency of 5 kHz. Labels in the figure denote light power ($\lambda = 532$). (d) Carrier density change produced by capacitive effect, calculated by $\Delta n_s = \frac{q s V_G}{e d}$ adopting the permittivity under a constant electrical field marked beside the curve and $V_G = 100$ V. Symbols are experimental values for $|V_G| = 100$ V extracted from literature, as indicated in the figure.
drop shown in Fig. 2c for $V_G < -20$ V and $P = 32$ mW, which suggests the exhaustion of sheet carriers. A large $\Delta n_S$ ($\sim 1.1 \times 10^{13}$ cm$^{-2}$ for a $V_G$ of $-200$ V) is also detected in illuminated c-LAO/STO (Supplementary Fig. 6). However, the illumination enhancement is almost absent when the interface is positively gated. In this case, as shown by the data of $V_G = 0$ (black) and 100 V (magenta) in Fig. 2a, $\Delta n_S$ increases slightly and can be ascribed to the illumination-generated extra photocarriers.

**Figure 3 | Field effect measured in different lights.** (a) Sheet resistance of a-LAO/STO corresponding to the field switching between on and off states, collected at a constant light power (32 mW) but different wavelengths. For clarity, only the data for $P = 0$ and $\lambda = 532$ nm are shown for $V_G = +40$ V. (b) Sheet resistance as a function of light wavelength, acquired at the time of 200 s for $V_G < 0$ and 1,000 s for $V_G > 0$. Solid lines are guides for the eye. All the measurements were conducted at room temperature.

**Figure 4 | Light illumination acceleration of the field-induced structural deformation of STO.** (a) Experiment set-up for the structural measurements of a-LAO/STO with simultaneously applied light illumination and gate field. (b) X-ray diffraction patterns of the 002 reflection of STO measured after a waiting time of 10 min upon the simultaneous application of light illumination ($P = 100$ mW, $\lambda = 532$ nm) and gate biases. The two shoulders developed on the low-angle side of the 002 reflection mark the lattice expansion in the near interface region of a-LAO/STO. Labels besides the curves indicate gate voltage. The total time required for each $\theta = 2\theta$ scanning is $\sim 10$ min. (c) A comparison of the lattice constants obtained with and without light illumination. The acceleration of the field-induced structural deformation by photoexcitation can be clearly seen. Solid lines are guides for the eye.
According to the capacitor model, the depleted carrier density by a negative $V_G$ is $\Delta n_p = \frac{\varepsilon_0 |V_G|}{\varepsilon_0 |V_G| + d}$, where $\varepsilon$ is the relative dielectric constant of STO and $d$ is the thickness of STO. Adopting the $\varepsilon$ values in ref. 17, the tuned carrier density by the capacitive effect can be calculated. As shown in Fig. 2d, the illumination-enhanced gating effect is much stronger than the simple gating effect that is always well described by the conventional capacitive effect. This result strongly suggests that additional mechanisms are at work under light illumination. To explore the origin of this unusual illumination-enhanced asymmetric gating effect, we examined the dependence of the field effect on light wavelength, $\lambda$. As shown in Fig. 3a, the tuned value of $R_s$ drops rapidly as $\lambda$ increases from 532 to 980 nm, suggesting that the photoexcitation of trapped electrons play a key role in the observed effect, although, counter-intuitively, the photoexcitation process significantly decreases, rather than increases, $n_p$. Furthermore, a strong-to-weak crossover of the illumination effect occurs at $\lambda$ ~ 850 nm (Fig. 3b, $V_G = -20$ V), corresponding to a photon energy of ~1.4 eV. This value coincides well with the reported deep oxygen vacancy states with one trapped electron in STO18,19.

Gating-induced and illuminating-enhanced lattice polarizations. As reported, oxygen vacancies ($V_O$) tend to pile up close to the STO surface20–23 and drift slowly under electrical field24. A recent study has shown that the electromigration of oxygen vacancies can lead to a polarity-asymmetric interface polarization25, which was built up in >20 h under a strong field. This slow buildup of the polar phase is reminiscent of the slow gating process observed when only the electrical field is applied (Fig. 1b). It is therefore possible that the illumination-enhanced gating effect is triggered by the acceleration of the establishment process of this interface phase. Direct evidence comes from Fig. 4, where a field-induced structural deformation of a-LAO/STO is indicated by X-ray diffraction. Figure 4a is the experimental set-up for structural measurements. Figure 4b is the $\theta-2\theta$ diffraction patterns, and Fig. 4c is the deduced out-of-plane lattice constant of STO, as a function of electric biases. When illuminated, as shown by Fig. 4b, a low-angle shoulder of the 002 reflection of STO emerges and develops above $V_G \approx -300$ V, indicating an out-of-plane lattice expansion. However, no structural changes are observed up to the gate bias of ~700 V without illumination. This result indicates that the light illumination indeed helps the gate field in inducing a structural deformation. As revealed by the previous work25, the lattice expansion of STO is a signature of interface polarization25. The structural distortion could not be a thermal effect as it continues to remain once it has appeared even after the sample is shaded from light, and the illumination alone produces no effect on structure (see Supplementary Fig. 7). Notably, the threshold $V_G$ for structural deformation is much higher than that for remarkable resistance tuning. It may be a consequence of uneven gating due to preferential carrier exhaustion around electrode, which confines gate field to the close proximity of the electrode. The uneven tuning can be sensed by sheet resistance since the latter is susceptible to local environment but not by X-ray diffraction unless $V_G$ is so high that the strongly gated area has well outward extension. We also performed the X-ray diffraction measurements for a Ti (30 nm)/STO/Ti (200 nm) capacitor structure and observed a similar lattice expansion (Supplementary Fig. 7). Without uneven gating, here the interface phase appears under a $V_G$ below −200 V. Corresponding to the emergence of interface phase, forbidden shifts in Raman spectra were detected, implying an inversion symmetry breaking (Supplementary Fig. 8). These results strongly suggest that light illumination has greatly accelerated the formation of the interface polarization phase.

Discussion
On the basis of the above analyses, we can present a scenario for the illumination-enhanced gating effect. As schemed in Fig. 5a,
the oxygen vacancy concentration at the LAO/STO interface is considerably high due to the outward oxygen ion diffusion from the STO substrate during the deposition of the LAO overlayer, and the resulted electron doping leads to the 2DEG at the α-LAO/STO interface and might also contribute to the conduction of the c-LAO/STO interface. Without illumination, negative bias only slightly polarizes the interface region of STO (Fig. 5b), yielding the slow Rg growth following the first sudden Rg jump (Fig. 1c). Light illumination accelerates interface polarization by enhancing the electromigration of oxygen vacancies, probably by exciting the trapped electrons in deep oxygen vacancy states (Fig. 5c). This polarization yields an extra tuning to nS, amplifying the gating effect.

Effect of light illumination on the electromigration of oxygen vacancies in STO can be identified from the transient leakage current before resistance degradation, a broad current peak will appear when the Vgb in the near region of the bridge reach cathode (Fig. 5b). As shown by the Supplementary Fig. 1, for the STO biased by a Vgb of −600 V, the current peak at t = 580, 115 and 816 min for the light power of 0, 40 and 100 mW, that is, illumination indeed accelerates the migration of oxygen vacancies. As revealed by ref. 25, the polarization will disappear in several seconds after removing the external field. This is consistent with our observation that Rg quickly drops back when the gate bias is removed (Fig. 1b). No additional tuning is observed under positive gate fields since there are no structural changes (Fig. 4b).

In conclusion, our present work has revealed a unique control of ferroelectric instabilities, pioneering a new avenue for the resistive switching effects markedly reduce (Fig. 5c). This polarization yields an extra tuning to nS, amplifying the gating effect.

Effect of light illumination on the electromigration of oxygen vacancies in STO can be identified from the transient leakage current recorded under a constant d.c. bias. As well established, prior to resistance degradation, a broad current peak will appear when the Vgb in the near region of the bridge reach cathode. As shown by the Supplementary Fig. 1, for the STO biased by a Vgb of −600 V, the current peak at t = 580, 115 and 816 min for the light power of 0, 40 and 100 mW, that is, illumination indeed accelerates the migration of oxygen vacancies. As revealed by ref. 25, the polarization will disappear in several seconds after removing the external field. This is consistent with our observation that Rg quickly drops back when the gate bias is removed (Fig. 1b). No additional tuning is observed under positive gate fields since there are no structural changes (Fig. 4b).

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N.P. provided the amorphous samples and undertook the XPS analysis. Y.W.X. and H.Y.H. provided the crystalline samples. Y. Li and J.W. characterized the sample via AFM. Y.S.C., Y. Lei and Y. Li performed the experiments for interface polarization. B.G.S. oversaw the project. All authors commented on the manuscript.

**Additional information**

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