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# Optically excited multi-band conduction in $LaAlO_3/SrTiO_3$ heterostructures

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The low-temperature resistance of a conducting $LaAlO_3/SrTiO_3$ interface with a 10 nm $LaAlO_3$ film decreases by more than 50% after illumination with light of energy higher than the $SrTiO_3$ band-gap. We explain our observations by optical excitation of an additional high mobility electron channel, which is spatially separated from the photo-excited holes. After illumination, we measure a strongly non-linear Hall resistance which is governed by the concentration and mobility of the photo-excited carriers. This can be explained within a two-carrier model where illumination creates a high mobility electron channel in addition to a low mobility electron channel which exists before illumination. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4790844]

A conducting interface[1] between band-insulating perovskites $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) displays a wide variety of physical phenomena, such as superconductivity,[2] magnetism,[3–8] and quantum oscillations with 2D character.[9–11] Several mechanisms are suggested to describe the origin of conductivity at the LAO/STO interface.[12–17] However, the relative contribution of each mechanism appears to depend on the LAO film thickness and on the LAO growth conditions such as substrate temperature, oxygen partial pressure, and the post annealing treatment; the complete picture of the involved mechanisms is not yet clear.[18] In particular, growing 5–10 layers of LAO on STO yields a metallic interface with relatively high mobility and low electron concentration,[19,20] whereas growing 26 LAO layers with the same conditions results in a low mobility, high concentration electron system with interesting magnetic properties.[3] Tuning the transport properties at such a complex oxide interface by modulating the carrier density with light can both contribute to the understanding of its physics and open new pathways towards oxide-based optoelectronic device applications. It has been shown previously that interface conductivity in oxide heterostructures can be tuned by light or by an electric field.[21–25]

In this letter we report our investigation of the interface of a LAO/STO sample with 26 monolayers of LAO, using low-temperature (4.2 K) magnetotransport experiments under selective illumination. Illuminating the sample with UV light of energy greater than the STO band gap results in a sharp and persistent decrease of electrical resistance. Using Hall effect measurements, we show that before illumination there is a single, low mobility electron conduction band and that the resistance drop on illumination can be explained by the creation of a parallel conducting channel containing optically excited high mobility electrons.

Our sample was grown by pulsed laser deposition (PLD) and has a 10 nm thick (26 unit cells) LAO film on a $TiO_2$-terminated single crystal STO [001] substrate (treatment described in Ref. 26). The LAO film was deposited at a substrate temperature of 850 °C and an oxygen pressure of $2 \times 10^{-3}$ mbar, using a single-crystal $LaAlO_3$ target. The growth of the LAO film was monitored using *in situ* reflection high-energy electron diffraction. After the growth, samples were cooled to room temperature in the deposition pressure.

The sample was mounted on a ceramic chip carrier, and electrical contacts were made with an ultrasonic wire-bonder, using aluminium wires. The magnetoresistance and Hall resistance were measured at 4.2 K in a van der Pauw geometry, using a standard low-frequency lock-in technique with an excitation current of 1 $\mu$A. The temperature dependence of the sample resistance shows a low-temperature upturn, similar to that observed previously by Brinkman *et al*.[3] The sample was illuminated with light from a broad-band Xe-lamp (LSB521), which was filtered using a 30 cm single grating monochromator (Acton-SP2300) as well as longpass and bandpass filters, to tune the excitation energy. The obtained quasi monochromatic light ($\Delta\lambda \simeq 3$nm) was brought to the sample via an optical fiber.

In Fig. 1, we show raw data of the sample resistance as a function of time during light illumination in the energy range between 1.44 and 3.65 eV. Each step corresponds to a constant illumination with a specific energy during 1 min (light "ON"). After illumination, we waited for 10 min to allow the resistance to reach a reasonably stable value (light "OFF"), before illuminating with the next photon energy. The influence of photons with different energy is clearly seen as a series of steps in the sample resistance. When the energy of illuminating photons is smaller than the band gap of STO (3.4 eV), a small resistance decrease is observed. This small change in the sample resistance can be due to the finite absorption of incident illumination by in-gap states present inside the STO band gap.[27,28] When the photon energy is approaching the band gap of STO, there is a gradual increase in the change of sample resistance. The largest drop in resistance (more than 50%) occurs when the photon energy

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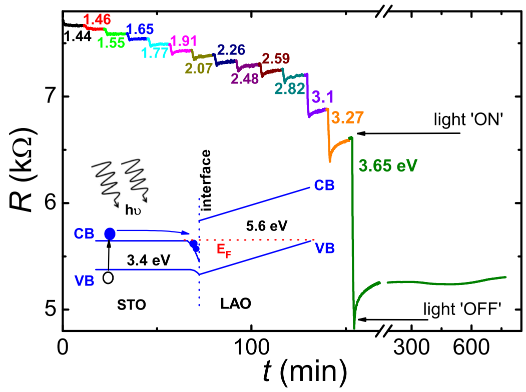


FIG. 1. Sample resistance as a function of time during the illumination with photons of energy from 1.44 to 3.65 eV at 4.2 K. Each change in the photon energy results in a pronounced step in the sample resistance; the photon energies, in eV, are shown beside each of the steps. Note the break on the time-axis 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 illumination and presuming an internal potential build up in the LAO.

(3.65 eV) exceeds the STO band gap energy (3.4 eV). This dramatically reduced resistance does not recover to the initial value after the illumination is turned off and only returns to the previous value when the sample is heated to room temperature. Repeated thermal cycling of the sample gave reproducible results.

We performed a control experiment on a STO[001] substrate, which was subjected to a similar chemical etching and annealing treatment (described in Ref. 26). The $TiO_2$-terminated bulk STO[001] substrate was heated to a temperature of 850°C at an oxygen pressure of $2 \times 10^{-3}$ mbar in the PLD chamber, without depositing the LAO layer. When illuminating this treated control sample, under similar illumination conditions as for the LAO/STO interface, we found that it remains insulating for all photon energies. This proves that the persistent resistance decrease is a feature of the LAO/STO interface and not an effect from the treated bulk STO[001] substrate.

In Fig. 2, we show as open circles (right axis), the variation in the number of illuminating photons as a function of photon energy, which is a consequence of the energy-dependent throughput of the optical set-up. The number of photons at the sample was calculated from the integration

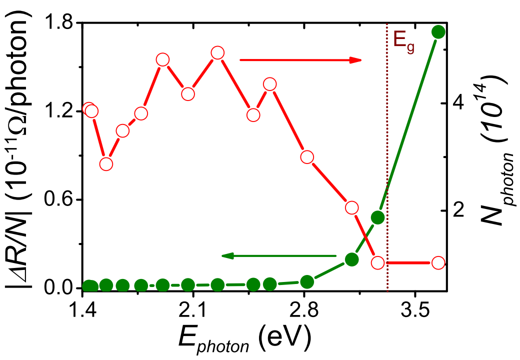


FIG. 2. Total number of photons at the sample as a function of photon energy is shown in open circles (right axis). Normalized sample resistance as a function of the energy of illuminating photons is shown in solid circles (left axis). The connecting lines are a guide to the eye.

over time of the total power incident on the sample, normalized with the energy of a single photon. The solid circles (left axis) show the resistance change of the sample, normalized by the number of incident photons, at each photon energy. The most drastic change occurs when the photon energy is higher than the STO band gap, shown as the vertical dashed line in the figure.

In order to explain these results, we propose the generation of additional, photo-excited carriers, as depicted in the inset of Fig. 1: Illuminating either the bulk STO (control sample) or the LAO/STO heterostructure with photon energy higher than the band gap of STO creates electron-hole pairs in the STO substrate. In the case of bulk STO, the photo-excited electrons in the empty conduction band do not persist for a long time and recombine very quickly with holes in the valence band, or through other recombination centres. For the case of LAO/STO heterostructure, the photo-excited electrons move to the interface potential well, where the holes remain trapped in the substrate. Owing to this spatial separation, electron and hole wavefunctions do not overlap, and direct optical recombination is suppressed, leading to a persistent resistance change.

For the LAO/STO heterostructure, the interface potential lifts the degeneracy of the STO bulk bands (Ti-3d xy, yz, and xz orbitals), and a further formation of 2D subbands is expected due to spin-orbit coupling and the internal electric field due to growth of LAO layers.[29] The 2D interface potential well gives a multi-subband character to the STO conduction band at the interface of LAO/STO.[11] Owing to this multi-subband structure of the interface, we propose that the photo-excited carriers occupy an initially unoccupied subband with a high mobility.

To study the nature of the persistent photo-excited carriers, we have performed magnetotransport experiments after illumination with an increasing total number of photons $N_{tot}$, controlled with neutral density filters, at a constant photon energy of 3.65 eV. In Fig. 3(a), we show the Hall resistance $R_{xy}$ as a function of applied magnetic field $B$ at 4.2 K, before illumination and after illuminating with four different values

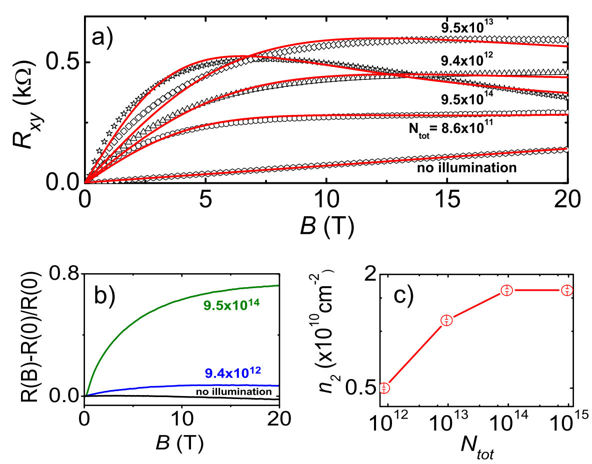


FIG. 3. (a) Hall resistance data as a function of the applied magnetic field, for illumination with different values of $N_{tot}$ with energy of 3.65 eV at 4.2 K (open symbols). Solid lines: The two-band model fits to the experimental data. (b) (longitudinal) Magnetoresistance data as a function of the applied magnetic field and (c) carrier concentration of the second, high mobility band for illumination with different values of $N_{tot}$.

of $N_{tot}$ (open symbols). The corresponding (longitudinal) magnetoresistance is shown in Fig. 3(b). Without any illumination, a linear Hall resistance and a small negative magnetoresistance are observed, in agreement with earlier observations on a similar sample.[3] After illumination, a distinctly non-linear Hall resistance and a large positive magnetoresistance appear.

We describe the linear Hall resistance using the conventional single-carrier model and extract the carrier concentration ($n = B/R_{xy}e$) and mobility ($\mu = 1/\rho_0 en$) from the slope of the linear fit to the data—the fit is shown as a solid line in Fig. 3(a), where $\rho_0$ is the zero-field sheet resistance and $e$ is the electronic charge. This yields a carrier density $n_1 = 8.9 \times 10^{13}\,\mathrm{cm}^{-2}$ and a mobility $\mu_1 = 3\,\mathrm{cm}^2/\mathrm{Vs}$. This very low carrier mobility is similar to values previously observed in LAO/STO samples with comparable LAO layer thickness.[3,30]

In contrast, the non-linear Hall resistance after illumination cannot be explained within a single-carrier model but rather suggests a multi-channel system. A similar non-linear Hall resistance was observed previously in $LaTiO_3$/$SrTiO_3$[31,32] and explained in terms of two-channel conduction from electronic bands with different mobilities, $\mu_1$ and $\mu_2$, and carrier densities, $n_1$ and $n_2$. We use a similar, simple two-electron-band expression for $R_{xy}$, given by[33]

$$R_{xy} = \frac{B}{e}\frac{(n_1\mu_1^2 + n_2\mu_2^2) + (\mu_1\mu_2 B)^2(n_1+n_2)}{(n_1\mu_1 + n_2\mu_2)^2 + (\mu_1\mu_2 B)^2(n_1+n_2)^2}, \qquad (1)$$

to model our Hall resistance data after illumination.

In this expression, we take $n_1$ and $\mu_1$ to be the carrier density and mobility of the existing electron band without illumination, and $n_2$ and $\mu_2$ are the carrier density and mobility of the persistent, photo-excited high mobility band. For low magnetic fields where $\mu_1 B \ll 1$ and $\mu_2 B \ll 1$ and for high magnetic fields where $\mu_1 B \gg 1$ and $\mu_2 B \gg 1$, expression (1) is linear in magnetic field; $R(B)$ becomes non-linear where $\mu_2 B \simeq 1$. This simple two-band model is not able to reproduce all the details of the magnetotransport in our sample, but the fact that we observe a non-linear Hall resistance at a few tesla clearly points towards the existence of an optically excited high mobility channel.

The results of our two-band analysis are shown in Fig. 3(a) as solid lines. We fitted the non-linear Hall resistance for $N_{tot} = 8.6 \times 10^{11}$, using $n_2$ and $\mu_2$ as fit parameters, and with fixed values of $n_1 = 8.9 \times 10^{13}\mathrm{cm}^{-2}$ and $\mu_1 = 3\,\mathrm{cm}^2/\mathrm{Vs}$ (as extracted from the linear Hall resistance before illumination): values of $n_2 = 0.5 \times 10^{10}\,\mathrm{cm}^{-2}$ and $\mu_2 = 1200\mathrm{cm}^2/\mathrm{Vs}$ were obtained. For the higher values of $N_{tot}$ we found that the quality of the fits was insensitive to small variations in $\mu_2$ around this value of 1200 $\mathrm{cm}^2$/Vs. We therefore fixed $\mu_2 = 1200\,\mathrm{cm}^2/\mathrm{Vs}$ and used only $n_2$ as a fit parameter for the higher values of $N_{tot}$. The values of $n_2$ extracted in this way are shown in the Fig. 3(c). For the highest value of $N_{tot}$, a good fit to $R_{xy}$ required a slightly increased value of $n_1$ from 8.9 $\times 10^{13}\,\mathrm{cm}^{-2}$ to $1.19 \times 10^{14}\,\mathrm{cm}^{-2}$, with an unchanged value of $n_2$ (as shown in Fig. 3(c)).

This two-band analysis of the Hall resistance strongly suggests that we populate a second, high mobility electron channel by illumination above the STO band-gap. This observed relatively high mobility (1200 $\mathrm{cm}^2$/Vs) of the persistently photo-excited electrons is similar to the values previously observed in reduced or non-stoichiometric bulk STO.[28,34–36] The fact that we see comparable high mobility values for photo-excited carriers suggests that the influence of interface defects is not so crucial in this case. A surface morphology study of the STO [001] substrate using an atomic force microscope, prior to growth of the LAO layer, did not reveal any macroscopic defects.

In summary, we have measured magnetotransport in a LAO/STO heterostructure, with a 10 nm LAO film, after illumination with selective photon energy. When the photon energy exceeds the STO band gap, the low-temperature resistance decreases by more than 50% and remains persistent at the lower value. We explain this effect in terms of optical excitation of an additional high mobility electron channel, which is spatially separated from the photo-excited holes, and confirm the presence of a second conducting electron band through measurement of a strongly non-linear Hall resistance after illumination. A two-carrier description of the Hall resistance data after illumination shows one low mobility (3 $\mathrm{cm}^2$/Vs) band with a high carrier density ($\simeq 10^{14}\,\mathrm{cm}^{-2}$) corresponding to the original conduction band present before illumination, and one persistently photo-excited high mobility (1200 $\mathrm{cm}^2$/Vs) band with a low carrier density ($\simeq 10^{10}\,\mathrm{cm}^{-2}$). We suggest that these persistently photo-excited carriers occupy one of the interface 2D subbands in STO, where these carriers have relatively higher mobility than the existing low mobility carriers without illumination.

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