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Modulation of Quantum Transport in Complex Oxide Heterostructures with Proton Implantation

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Abstract:

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The interfacial electronic properties of complex oxides are governed by a delicate balance between charge transfer, lattice distortions, and electronic correlations, posing a key challenge for controlled tunability in materials research. Here, we demonstrate that proton implantation serves as a precise tool for modulating interfacial transport in SrTiO₃-based heterostructures. By introducing protons into the SrTiO₃ substrate beneath an amorphous (La,Sr)(Al,Ta)O₃ capping layer, we uncover a competition between disorder and charge doping induced by implantation. At low implantation fluences below 1×10^{15} protons/cm² (1E15), charge doping dominates, leading to an increase in carrier density and mobility, analogous to electrostatic gating effect. This enables the emergence of quantum transport oscillations at low temperature. Conversely, at higher fluences (above 1E15), disorder scattering prevails, suppressing carrier mobility and inducing an insulating state. The nonmonotonic evolution of transport with implantation fluence underscores the critical interplay between electronic correlations and disorder, offering a new paradigm for the controlled engineering of interfacial quantum states in SrTiO₃-based oxide heterostructures.

Keywords: proton implantation, oxide heterointerface, two-dimensional electron system, structural damage, charge doping

Introduction

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Continuous effort has been made to develop the technique of ion implantation to fabricate commercial semiconductor devices since 1957.^{1–3} One example is the widely-used silicon-on-insulator substrate, which can be obtained using oxygen ion implantations followed by a high-temperature annealing.⁴ Another example is the smart-cut process, where the high-dose ion implantation is applied to create a cracking layer at a specific location to induce an in-depth splitting in the target sample.⁵ Moreover, it has been demonstrated that over 40 steps of ion implantations, with various doses and energies, are required to achieve a modern 28-nm “system on a chip” device.⁶ So, ion implantation has played an important role in developing novel functionalities and device fabrications in Si-based industry.² On the other hand, the oxide heterointerface is capable of integrating multiple functionalities into one device and has been proposed as a possible solution to preserve Moore’s law in future.⁷ So, it is curious to clarify whether the ion implantation, a fully-developed technique in modern semiconductor industry, can be applied to functional oxide heterointerfaces for designing the next-generation electronic devices.

A good example of functional oxide heterointerface is the SrTiO₃-based interface, where multiple properties including the two-dimensional (2D) conductivity,^{8,9} magnetism,^{10,11} superconductivity,¹² ferroelectricity¹³ and spin-orbital coupling^{14–16} are coexisting. There are several reports investigating the ion-implantation effect on the well-known conducting LaAlO₃/SrTiO₃ interface. Mathew *et al.* used 2 MeV protons with dose above 6×10^{17} protons/cm² (6×10^{17}), or 500 keV He ions with dose above 1×10^{16} , to remove the interfacial conductivity of exposed areas.¹⁷ Similarly, Hurand *et al.* applied oxygen ions (50 keV, 5×10^{12} cm⁻²) to pattern the LaAlO₃/SrTiO₃ interface for obtaining the top-gated field-effect transistor, of which the micro-size channel protected from the ion implantation maintains the metallic transport behavior.¹⁸ Also, Aurino *et al.* studied the post thermal annealing, which heals the

ion-implantation-induced damages to recover the interfacial conductivity.^{19,20} All those studies focus on the ion-implantation-induced *structural damage*, which creates disorders for carrier localizations at the ion-implanted SrTiO₃-based interface. However, the other side of ion implantations, *charge doping*, at oxide heterointerfaces, is not fully discussed. During the ion implantation, the high-energy ions will knock out the oxygen in oxides, leaving oxygen vacancies (as localized positive charges) and excited electrons (as mobile negative charges) in SrTiO₃. It has been well documented that the insulating SrTiO₃ can be easily turned into a conductor by various types of electron doping, including chemical substitutions or electrostatic gating.²¹ In this work, we will present and discuss about two sides of the ion-implantation effect, *structural damage* and *charge doping*, which simultaneously affect the SrTiO₃-based interface.

Results and discussions

We used 50 keV protons (or H⁺ in some figures) for ion implantation, and the target oxide heterointerfaces are prepared by growing the amorphous (La,Sr)(Al,Ta)O₃ (*a*-LSAT) layer on the proton-implanted (001) SrTiO₃ substrate with different implantation doses. If implantation were performed after deposition, the implanted protons would traverse the already formed conducting interface and severely disrupt it, rendering the interface insulating. **Figure 1(a-c)** summarize the process of sample preparation. First, the SrTiO₃ substrate was treated by the buffered HF and thermal annealing to achieve an atomically flat TiO₂-terminated surface. Second, the protons were implanted into the treated SrTiO₃ substrate with different doses, ranging from 1E14 to 1E16. **Figure 1(d)** presents the gradual change of colors in proton-implanted SrTiO₃ substrate. When the virgin SrTiO₃ substrate (without proton implantation) is colorless and transparent, the color becomes darker and opaque with the higher implantation dose. This is because the proton implantation produces oxygen vacancies, accompanied by the formation of in-gap states to enhance the absorption of visible lights in the darkened SrTiO₃.²² Although those implanted SrTiO₃ substrates contain some oxygen vacancies, they still maintain

the insulating nature with resistance $R > 10^8 \Omega$. Third, the *a*-LSAT layer was grown on the proton-implanted SrTiO₃ substrate by pulsed laser deposition (PLD) under the high-vacuum and room-temperature condition. The high vacuum is required for the formation of oxygen-vacancy-induced quasi-two-dimensional electron system (*q*-2DES) at the amorphous SrTiO₃ heterointerface, and the room-temperature deposition is adopted to avoid the high-temperature process that could compromise the ion-implantation effect.²³ Therefore, the proton-implanted *a*-LSAT/SrTiO₃ sample is expected to consist of two important charged regions: one is the conventional oxygen-vacancy-induced *q*-2DES close to the heterointerface (red region in **Figure 1(c)**), and the other one is the implanted SrTiO₃ layer (green region in **Figure 1(c)**) that is far away from the heterointerface and contains implanted protons with resulted defects.

Figure 2 summarizes the basic transport properties of *q*-2DES at the proton-implanted *a*-LSAT/SrTiO₃ interfaces. To emphasize the modulation of proton-implantation in **Figure 2(a)**, the temperature-dependent sheet resistances, obtained from samples with different proton doses, which are normalized with respect to that of the virgin sample (without proton implantation) as $R_{imp}(T)/R_{vir}(T)$. The nonnormalized temperature-dependent sheet resistances of the virgin and implanted samples are provided in **Figure S1**. The room-temperature sheet resistances (measured at 300 K) are monotonically reduced on increasing proton fluence from 0 to 5E15. However, the low-temperature sheet resistances (measured at 2 K) don't follow this monotonical trend: the low-temperature resistances reach the minimal value when proton fluence is around 1E15. Further increasing the implantation fluence rapidly raises the low-temperature sheet resistances, accompanied by a transition from the metallic behavior ($dR/dT > 0$) to semiconducting ($dR/dT < 0$). Moreover, the sheet resistances are finally out of our measurement range ($R > 10^8 \Omega$) when the proton fluence is above 1E16, indicating an insulating behavior. In **Figure 2(b)**, the room-temperature (300 K) and low-temperature (2 K) carrier densities n_s are plotted as a function of proton dose. Our results reveal a clear proton-

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implantation-induced enhancement on n_S , even in the high-fluence samples (up to 5×10^{15}) with semiconducting behaviors. Given that the implanted SrTiO₃ substrate is not conducting without the on-top *a*-LSAT layer, the observation of enhanced n_S suggests a strong interaction between two charged regions – the proton-implanted SrTiO₃ layer and *q*-2DES interface. Also, carrier mobilities μ_S measured at room temperature and low temperature are compared in **Figure 2(c)**. While the room-temperature μ_S is almost constant around $3\text{--}8 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, the low-temperature μ_S is very sensitive to the proton fluence. The low-temperature μ_S reach the maximum value $\sim 10,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ with the proton fluence around 1×10^{15} , corresponding to the minimal low-temperature R . Hence, the suppression on metallic behavior in high-dose *a*-LSAT/SrTiO₃ heterointerfaces is caused by the reduction on μ_S rather than n_S .

To investigate the location of proton-implanted layer in the SrTiO₃ substrate, **Figure 3(a)** presents the simulation results performed by Stopping and Range of Ion in Matter (SRIM).²⁴ According to the SRIM results, the end of range is at around 300 nm underneath the surface, the proton distribution is a bit deeper than the vacancy region. A detailed SRIM result about vacancy creation is shown in **Figure S2**, which suggests that most of the vacancies are oxygen vacancies.

Figure 3(b) compares the ω - 2θ scans of X-ray diffraction (XRD) obtained from samples with different proton fluences. While the (002) peaks (indexed by a dash line) that represent the unaffected part of SrTiO₃ is unchanged on increasing the proton dose, the left-side shoulders (indexed by a solid line) resulted from the proton-implanted SrTiO₃ layer with defects become significant. Also, those left-side shoulders reveal the lattice expansion of proton-implanted SrTiO₃, which can be ascribed to the formation of oxygen vacancies as discussed above.^{22,25–30} On the other hand, the cross-section image obtained from transmission electron microscopy (TEM) reveals that the implanted SrTiO₃ layer is ~ 450 nm away from the *a*-LSAT/SrTiO₃ heterointerface, as shown in **Figure 3(c)** and **Figure S3**. The actual damage

depth is deeper than simulation result. This might be because of the channeling effect of proton beam in the crystal lattice. Nevertheless, the fact that proton-implanted SrTiO₃ layer is located well below the *q*-2DEG layer is identified. Meanwhile, there are limited number of disorders created at the *a*-LSAT/SrTiO₃ interface during the proton implantation to affect the *q*-2DES. It is expected that when the implantation fluence is high enough, the *structural-damage*-induced disorders will raise the energy position of mobility edge with respect to the Fermi level (E_F , Fermi energy), leading to Anderson-localization to remove the 2D conductivity at the interface.^{31–33} The non-monotonic mobility can be rationalized within an Anderson-localization framework in which extended states exist only for energies above a disorder-dependent mobility edge energy E_C .^{34–36} By combining Hall densities with the Poisson–Schrödinger Fermi energies and fitting the low-temperature conductivity to $(E_F - E_C)^{\nu}$, we find that E_C overtakes E_F near a fluence of 1E15 (see Supplementary Figure S6), coincident with the observed collapse of carrier mobility.

Given the above experimental results, we proposed a model that describes the charge distribution in the proton-implanted *a*-LSAT/SrTiO₃ interface as sketched in **Figure 3(d-e)**. When protons are implanted into a bare SrTiO₃ substrate, oxygen vacancies (O_V) are formed to ionize the positively-charge in-gap states ($O_V^{\bullet\bullet}$) and electrons (e^-) at the proton-implanted region. Because of the surface-depletion-induced band bending as shown in **Figure 3(d)**, the thermally excited electrons will be easily trapped by the defect state with $O_V \rightleftharpoons O_V^{\bullet\bullet} + 2e^-$, leading to the insulating property of the proton-implanted SrTiO₃ substrate. If the SrTiO₃ surface is covered by the *a*-LSAT or *a*-LaAlO₃ layer, the surface band will bend in an opposite way to create a potential well for *q*-2DES at the heterointerface as plotted in **Figure 3(e)**. In this case, electrons that are thermally excited from the defect states to conduction band will flow to the heterointerface with $O_V \rightarrow O_V^{\bullet\bullet} + 2e^-$. To substantiate the band-bending model, we performed a self-consistent Poisson–Schrödinger estimate (Supplementary Information). The

calculated Fermi energy increases from ~ 0.05 eV in pristine interfaces to ~ 0.3 eV at optimal H^+ dose, while the characteristic ground-state confinement length remains $\sim 6\text{--}10$ nm. The higher Fermi energy permits occupation of excited sub-bands, broadening the overall electron distribution and supporting the charge-transfer mechanism proposed in Figure 3(d-e). This is consistent with our observation that both the high-temperature and low-temperature n_S increase on the proton implantation. Hence, two sides of the proton-implantation effect, including *structural damage* and *charge doping*, are presented in the proton-implanted a -LSAT/SrTiO₃ heterointerface. While *charge doping* plays an important role in low-fluence samples ($\leq 1E15$), the effect of *structural damage* becomes dominant on increasing the fluence ($\geq 1E16$).

Given that the ion-implantation-induced *structural damage* with high implantation dose has been well reported,^{17–19} we focus on the effect of *charge doping* in low-dose samples. As shown in **Figure 4 (a, b)**, if the bottom implanted SrTiO₃ layer acts as the positively-charged donor and the top q -2DES as the acceptor with negative charges, the proton-implantation-induced charge doping can be mimicked by the back-gating electrostatic doping, where additional electrons are doped into the top q -2DES layer by applying a positive back-gating voltage. In **Figure 4(c)**, the relationship between low-temperature n_S and μ_S is revealed in the proton-implanted (with fluence no more than $1E15$) and back-gated a -LSAT/SrTiO₃ heterointerfaces. A consistent trend is observed in both cases, where the low temperature μ_S are improved by increasing n_S . One possible explanation is that the increased n_S enhances the screening effect to suppress the disorder-induced scattering. Another possible mechanism is that the positive charges (due to proton implantation) or voltage (from back-gating) underneath the q -2DES layer can draw the mobile electrons away from the interfacial defects by Coulomb interaction. Both effects mentioned above may effectively increase carrier mobilities by raising carrier densities. The similar modulation on carrier mobility, mediated by the low-dose proton

implantation and positive back-gating voltage, indicates the similar physics of charge doping in both methods.

By modifying the fluence of implanted proton, the carrier mobility of q -2DEG at the LSAT/SrTiO₃ interface is improved from 1,000 to 10,000 cm²V⁻¹s⁻¹. **Figure 5** presents low temperature magneto-transport properties of the selected proton-implanted sample, of which the proton fluence is 1E15 with n_S of 1.12×10^{14} cm⁻² and μ_S of 8,000 cm²V⁻¹s⁻¹. When temperature is around 2-3 K and magnetic field B above 6 T, the sample shows Shubnikov-de Haas (SdH) effect featured by the oscillating magnetoresistance in **Figure 5(a)**. If plotting the low-temperature MR as a function of $1/B$, the oscillating periodicity is around 0.017 T⁻¹. The density of high-mobility electron (n_{SDH}) that induces the SdH oscillations can be estimated by $n_{SDH} = \frac{2e}{h} \sum f_i$, where f_i frequencies compose the quantum oscillations. Accordingly, n_{SDH} is $\sim 7.5 \times 10^{12}$ cm⁻² which is much smaller than n_S obtained from Hall measurement. The ratio n_{SDH}/n_S ($\sim 0.1 - 0.3$) falls within the range as reported in the previous works,³⁷⁻⁴¹ indicating that only the light, high-mobility pockets contribute to the oscillations while the heavier or strongly scattered bands dominate the Hall signal. Such phenomenon with $n_{SDH} < n_S$ is widely observed in the high-mobility q -2DES at the SrTiO₃-based heterointerface, probably due to the complicated sub-band structure associated with multiple conducting channels. It is also clear to observe that the oscillation longitudinal resistance (ΔR) decreases with increasing temperature as shown in **Figure 5(b)**. The oscillation longitudinal resistance (ΔR) as a function of temperature (**Figure 5(c)**) can be defined as $\Delta R(T) = 4R_0 e^{-\alpha T_D} \alpha T / \sinh(\alpha T)$, where $\alpha = 2\pi^2 k_B / \hbar \omega_C$, $\omega_C = eB/m^*$, k_B is the Boltzmann constant, $\hbar$ is the Planck constant, ω_C is the cyclotron frequency, e is the elementary charge, B is the magnetic field, m^* is the carrier effective mass, R_0 is the non-oscillatory component of R_S , and T_D is the Dingle temperature. The fitting of these data by using the equation gives the effective mass $m^* = 0.95 \pm 0.04 m_e$,

where m_e is free electron mass and Dingle temperature $T_D = 2.4 \pm 0.3$ K. This m^* value is consistent with a moderately renormalized t_{2g} band at the a -LSAT/STO interface.^{37,42–45}

To sum up, we have shown that structural damage and charge doping—two different directions of the ion-implantation effect are both existing. An optimum proton-implanted (1×10^{15} for 50 keV proton) a -LSAT/STO sample can lead the high carrier mobility which enables quantum transport oscillations at low temperature. On the other hand, samples with high implant fluences (more than 1×10^{15} protons/cm²) show signs of structural damage, which leads to reduced carrier mobility and insulating behavior. This offers a practical method for adjusting transport properties at SrTiO₃-based conducting interfaces in oxide heterostructures, opening avenues for exploring innovative functionalities.

Materials and Methods

Sample preparation. The 0.5 mm thick (001) SrTiO₃ (STO) substrate (Crystec) was treated with HF and annealed to obtain defined terrace steps and TiO₂-terminated surface. Substrates with proton-implantation were transferred to ion irradiation accelerator prior to the amorphous LSAT (*a*-LSAT) deposition. The pulsed laser deposition method was used for sample preparation. *a*-LSAT was grown at room temperature and high vacuum (10⁻⁶ Torr). During the growth, a nanosecond KrF 248 nm laser was used with a fluence of 2.0 J cm⁻² and a repetition rate of 2 Hz.

Ion irradiation. A SingletronTM accelerator was used to generate H₂⁺ ion beams from a hydrogen source bottle and 100kV terminal voltage. 100keV H₂⁺ was selected by controlling a 90-degree magnetic field. The beam was focused with a quadrupole lens set to a spot size about 50μm × 50μm, and scanned over the whole sample.⁴⁶ The irradiation fluence was controlled by the beam current and irradiation dwell time at each pixel.

Electrical measurements. Sheet resistance, carrier densities, and carrier mobility were determined using the Van der Pauw method on a physical property measurement system (Quantum Design), which allowed for precise characterization of the electrical properties of the samples. Magneto-transport measurements were conducted over a broad magnetic field range, up to 9 Tesla, to assess quantum oscillations phenomenon on the transport measurements.

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Data availability

The data that support the findings of this study are available from the corresponding authors upon request.

Main text Figures

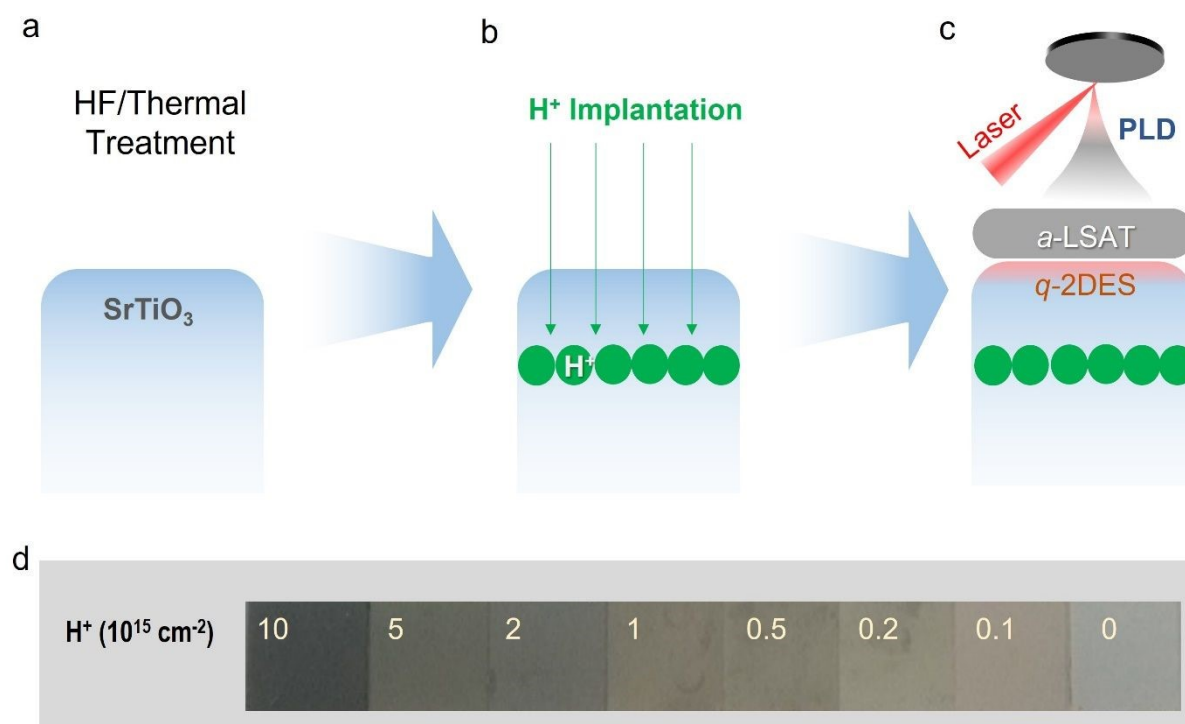
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Figure 1. Schematic of the sample preparation and optical images of the samples before and after proton implantation. (a) Buffered HF and thermal annealing treatment with the STO wafer. (b) Schematic of proton implantation in the treated STO. (c) PLD of a-LSAT process on the treated and implanted STO wafer. (d) Optical images of the STO before (right most, marked as 0) and after proton implantation with different doses from $1\text{E}14$ to $1\text{E}16$ (from right to left).

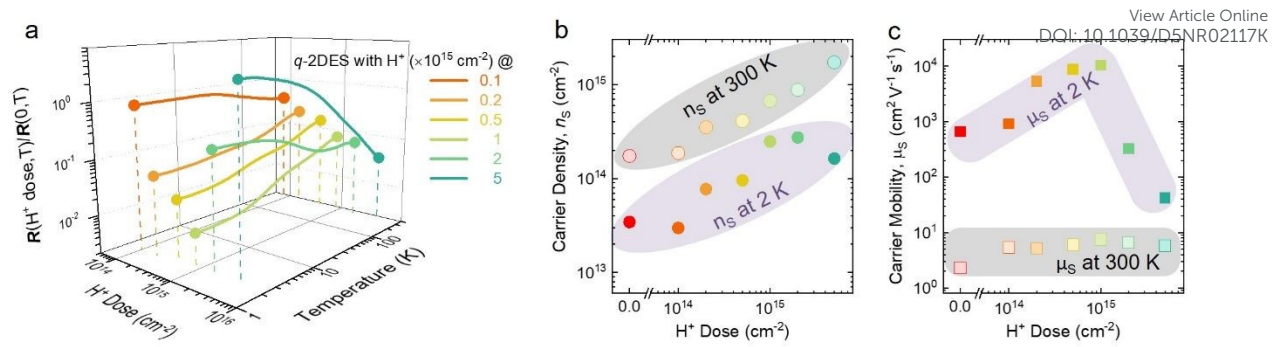


Figure 2. Transport properties of the *a*-LSAT/SrTiO3 interfaces. (a) Relative resistances the proton implanted samples comparing with the virgin sample. (b) Carrier densities and (c) mobilities of the virgin and implanted samples at 2K and 300K.

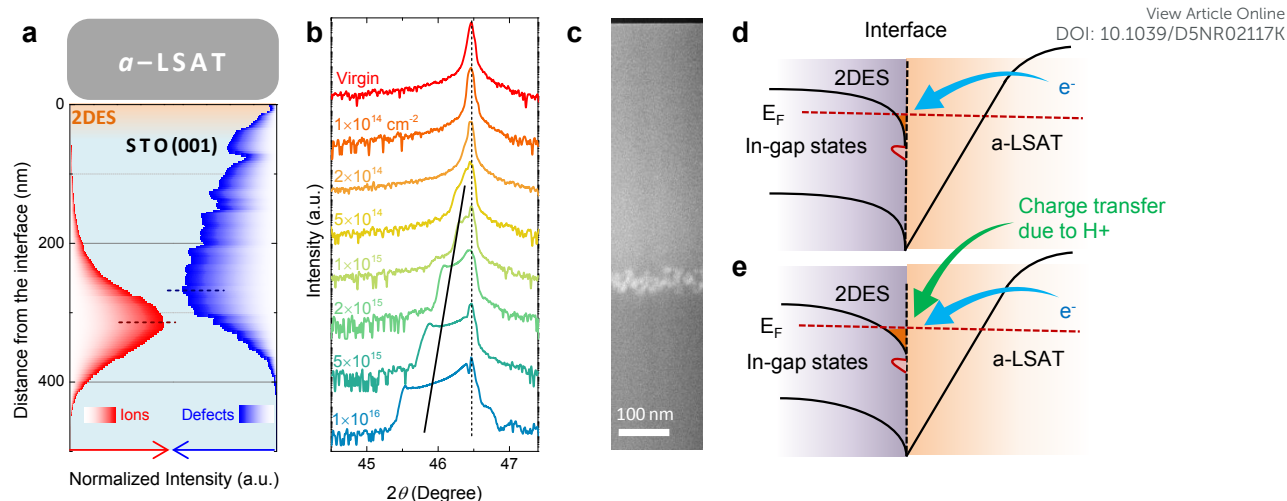


Figure 3. Ion and vacancy distribution of the proton implanted STO wafer. (a) SRIM result of 50 keV proton in STO. (b) XRD of the STO before and after proton implantation. (c) TEM image of the cross section of the irradiated STO. (d) Band bending model of *a*-LSAT/STO with in-gap states with low carrier density. (e) and with higher carrier density due to H⁺ charge transfer.

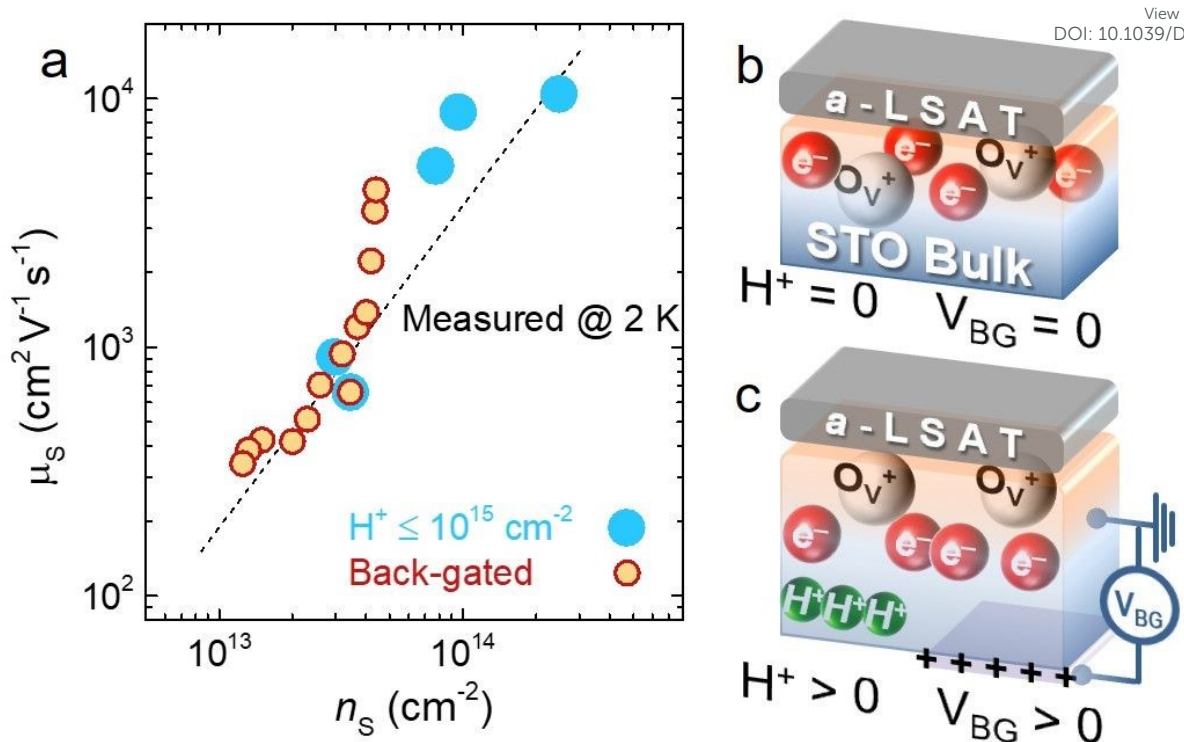


Figure 4. Comparison of the proton implanted and back-gated induced transport. (a) Relationship between μ_s and n_s at 2 K for the ion implanted with back-gated induced transport. Schematics to show the difference in STO electron carrier density in (b) virgin and (c) implanted samples with back-gate.

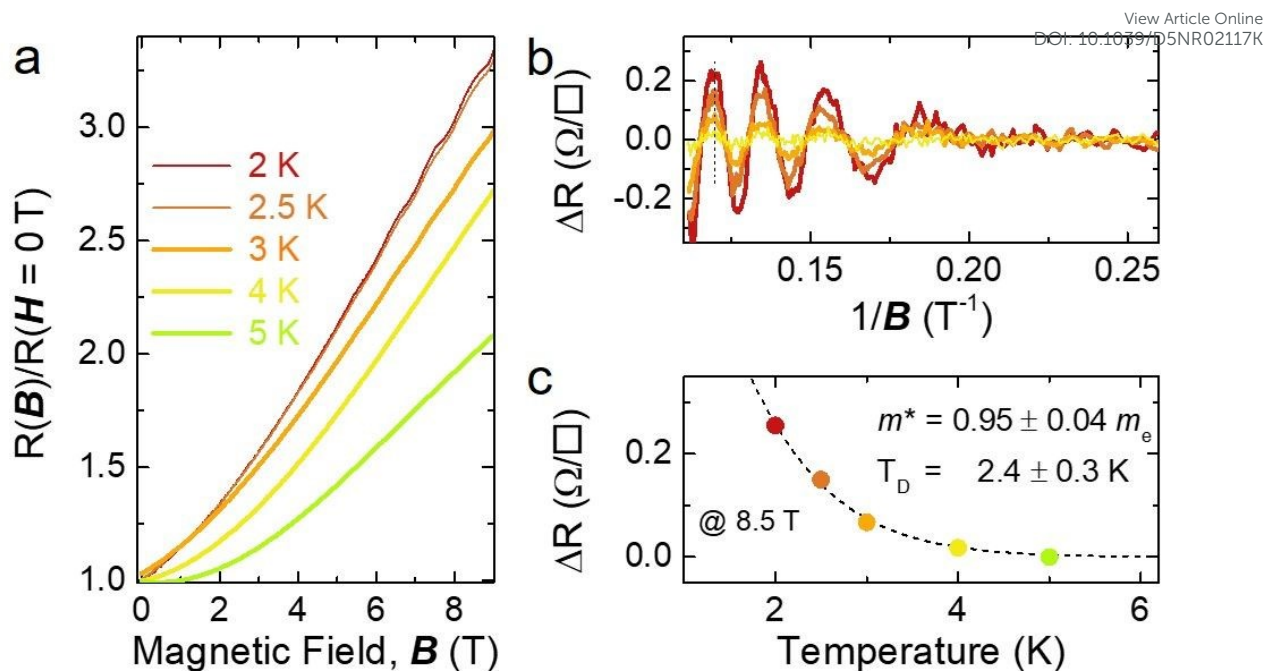


Figure 5. Low temperature magneto-transport properties. (a) Longitudinal resistance of the selected proton-implanted sample as a function of the field for different temperature ranging from 2 K to 5 K. (b) Inverse-field dependence of the oscillating longitudinal resistance (ΔR). (c) Temperature dependence of the longitudinal resistance (ΔR) for 8.5 T magnetic field. Symbols are the experimental data, and the solid lines are the Lifshitz-Kosevich (L-K) fit. Note: Figures 5(b) and 5(c) follow the same colour scheme as indicated in the legend of Figure 5(a). Light green colour plot (5K) in Figure 5 (a) doesn't have any oscillations, therefore it is not shown in the Figure 5 (b).

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Data availability

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The data that support the findings of this study are available from the corresponding authors upon request.