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Anatase TiO₂-based two-dimensional electron gases generated by low-energy argon-ion irradiation

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Two-dimensional electron gas (2DEG) has abundant properties, which motivate not only fundamental physical interest but also concepts for device designing. While most of the reported 2DEGs are based on SrTiO₃, here we report on a systematic investigation on anatase TiO₂-based 2DEG generated by ion irradiation. By irradiating the surface layer of the anatase TiO₂ films with appropriate argon ion beams, we gained the 2DEGs with a thickness of 4 nm. Unique transport behaviours of a $T^{-1/3}$ or ln(1/*T*) sheet resistance dependence below 100 K are observed depending on irradiation time. Unusually large positive MR is observed for which the magnetic field causes a strong shrinkage of electron wave functions, and fairly large negative MR is detected in the 2DEG, arising from the depression of magnetic scattering. *Published by AIP Publishing*. https://doi.org/10.1063/1.5030081

Two-dimensional electron gases (2DEGs) at oxide interfaces have attracted extensive attention because of their wealth of unique physical properties,^{1–3} providing opportunities for the development of the next generation of electronic and photonic devices.⁴ There are several approaches that can lead to oxide 2DEGs, such as charge transfer,⁵ δ -doping,⁶ or oxygen vacancies (V_Os). Among them, the latter approach is the most feasible way.

Besides growth conditions, high-temperature reduction, chemical doping, ion-beam implantation, or noble-ion irradiation can also generate Vos and even modify their spatial distribution. Compared to vacuum annealing, ion beam irradiation is a unique approach due to its ability to control the distribution of V_Os in a predetermined depth by an appropriate choice of ion species, energy, and dose.^{7,8} Moreover, ion irradiation can be used to tailor the structure and properties of nanometre-sized materials with high spatial precision,9,10 which offers more opportunities for the design of nanostructures with functionalities. As has been demonstrated,¹¹ after an appropriate argon ion (Ar⁺) irradiation, 2DEG can be formed in the surface layer of SrTiO₃ (STO). The 2DEG obtained here thus has essentially the same characters as those of the 2DEG at the STO-based heterointerfaces but can be feasibly tuned by irradiation dose.

TiO₂ is similar to STO in many aspects, such as the easy redox reaction and the high electronic conduction when oxygen vacancies exist.¹² But TiO₂ is unique due to its nontoxicity, chemical stability, and low production cost and has been widely used in many fields.^{13–15} Electronic and atomic reconstructions, especially lattice defects such as V_Os, can dramatically alter the properties of the TiO₂ surfaces,^{16,17} enabling many applications. Despite the intensive works on TiO₂, TiO₂-based 2DEG has been scarcely studied. Only a

few TiO₂-based 2DEGs were reported, and all of them were obtained at the oxide TiO₂ interfaces.^{18,19} There are no attempts to build up the 2DEG at the TiO₂ surface through ion irradiation. In this work, we fabricated the 2DEGs by irradiating the surface of anatase TiO₂ films on (001) LaAlO₃(LAO) substrates with Ar^+ ions. Unique transport behaviours are observed, accompanied by the one- to two-dimensional transition of the electron gas with irradiation time. The low-temperature magnetoresistance (MR), recorded in perpendicular fields, is initially positive and yet unusually large and then fairly negative. This work demonstrates the potential of Ar^+ irradiation in fabrication of well spatially defined low dimensional electron systems.

Anatase TiO₂ thin films were grown on (001)-orientated LAO crystals $(5 \times 5 \times 0.5 \text{ mm}^3 \text{ in dimension})$ because of their small lattice mismatch (0.18%). The thin films were prepared by pulsed laser deposition (PLD) using a KrF Excimer laser (wavelength = 248 nm) with a laser fluence of 1.5 J/cm^2 and a repetition rate of 2 Hz. A sintered TiO₂ (99.99%) pellet was used as the target. The LAO (001) substrates were degreased in ethanol and cleaned with deionized water and acetone with supersonic waves for 15 min. During deposition, the oxygen pressure was kept at 5×10^{-2} Pa and the temperature was fixed to 680 °C. The film thickness was \sim 36 nm, controlled by deposition time and subsequently confirmed by X-ray reflectivity (XRR, D8 Discover, Cu Ka radiation, Bruker). The Ar⁺ irradiation was done by an ion beam etching system (LKJ-1D-150) with an acceleration voltage of 200 V at a rate of 2.5×10^{17} ions/cm² s and an incident angle of 60°. The irradiation time varied from 0.5 to 3 min. The surface morphology of the films was measured using an atomic force microscope (AFM) (SPI 3800N, Seiko), and the crystal structure was determined using an X-ray diffractometer with a Cu Ka radiation (D/Max-2400, Rigaku). The X-ray photoelectron spectroscopy (XPS) measurements were performed in a Thermo Fisher ESCALAB

250 Xi system with a monochromatic Al Ka X-ray source (photon energy 1486.7 eV). The angle of the incident X-ray was varied from 0° to 55°. Resistance of the irradiated films was measured using a Quantum-Design physical properties measurement system (PPMS) in the temperature range from 2K to 300K and the field range up to 7T, with an applied current of 10 μ A. Ultrasonic wire bonding (Al wires of $20\,\mu\text{m}$ in diameter) was used for electric connection, and the four-point Van der Pauw geometry was adopted. The film surface is atomically flat. Figures 1(a) and 1(b) show the surface morphology of the TiO₂ films. The root-mean-squared (RMS) roughness of the films, measured over a $5 \times 5 \ \mu m^2$ area, is about 5.7 Å before irradiation and about 5.1 Å after a 2-min-irradiation. Figure 1(c) exhibits the XRR data of the film irradiated for 0.5 min. The thickness of the film is derived from the simulation of the experimental curve. Other three samples were analysed following the same procedure. The film thickness decreases from originally 36 nm to 23 nm after a 3-min irradiation, reducing at a rate of 4.3 nm/min. Figure 1(d) is a comparison of the X-ray diffraction (XRD) patterns of the film before and after ion irradiation. Welldefined XRD peaks, which are the (004) Bragg reflections of the anatase TiO₂ phase, appear at 37.96°, irrespective of Ar^+ irradiation, corresponding to an out-of-plane lattice constant of 9.47 Å that is close to the reported value of 9.51 Å. The full width at half maxima (FWHM) of the TiO₂ (004) peak are 0.29° and 0.37° before and after irradiation. All these results indicate that the anatase TiO₂ films are of epitaxial growth, with a (001) orientation.

 Ar^+ irradiation will introduce oxygen vacancies into TiO₂, thus reducing the valence state of the Ti ions. To confirm the formation of V_Os and reveal their depth distribution, the spectra of angle resolved X-ray photoelectron spectroscopy



FIG. 1. (a) and (b) Surface morphologies of the anatase TiO_2 films before and after an Ar^+ irradiation of 2 min, respectively. (c) XRR spectrum of the anatase TiO_2 film irradiated for 0.5 min (symbols) along with the results of data-fitting (red curve). (d) X-ray diffraction spectra of the anatase TiO_2 film before and after an argon-ion irradiation of 2 min. The XRD spectrum of the as-prepared sample was upwards shifted for clarity.

(XPS) were measured. As an example, in Fig. 2(a), we show the XPS spectra of the Ti $2p^{3/2}$ state after an irradiation of 3 min, measured at an emission angle of 0° (perpendicular to the film plane). The main Ti $2p^{3/2}$ peak at the binding energy of 458.7 eV is associated with Ti⁴⁺ ions, while the identifiable shoulder at 456.9 eV comes from Ti³⁺ ions.²⁰ Provided that the contents of the Ti³⁺ and Ti⁴⁺ ions are proportional to their respective integrated peak intensities, the Ti³⁺/(Ti³⁺ + Ti⁴⁺) ion ratio can be determined from the results of data fitting by the commercially available *Thermo Avantage software*. Figure 2(b) presents the Ti³⁺/(Ti³⁺ + Ti⁴⁺) ratio as a function of irradiation time. It is originally 0 and 0.13 after a 3-min irradiation. Similar spectra except for minor variations in the low-binding energy peak are obtained for other two emission angles of 30° and 55°.

Based on the XPS spectra collected at different emission angles, a quantitative description of the spatial distribution of the Ti^{3+} ions can be obtained adopting a simple model^{21,22}

$$I(Ti^{3+})/I(Ti^{3+} + Ti^{4+}) = p[1 - \exp(-t/\lambda\cos\omega)], \quad (1)$$

where $I(Ti^{3+} + Ti^{4+})$ is the integrated intensity of the Ti^{3+}/Ti^{4+} ions, ω is the emission angle, p is a constant fraction of Ti^{3+} per TiO_2 unit cell, t is the depth of the irradiation, and $\lambda = 2.3$ nm is the electron escape depth in TiO_2 .²³ Obviously, when the incident X-ray forms a larger angle with the film normal, it will transport for a longer distance in the interfacial layer, which is equivalent to the decrease in the effective electron escape depth ($\lambda \cos \omega$), thus exciting more Ti^{3+} ions that reside in the interfacial layer. This means that a monotonic growth of $I(Ti^{3+})$ is proportional to ω .

Best fitting of the $Ti^{3+}/(Ti^{3+} + Ti^{4+})-\omega$ relation to Eq. (1) gives rise to a p value ranging from 0.08 to 0.18 for the irradiation time from 0.5 to 3 min, increasing with irradiation time as expected, and the irradiation depths of 3.5 nm, 3.3 nm, 3.0 nm, and 3.2 nm corresponding to the irradiation time of 0.5, 1, 2, and 3 min, respectively. Obviously, the surface layer within which most of the Ti^{3+} ions reside is around 4 nm, revealing a quasi-two-dimensional distribution of the V_{OS} in the irradiated films. Notably, the thickness of



FIG. 2. (a) Ti $2p^{3/2}$ XPS spectra after an irradiation of 3 min at an emission angle of 0°. Red and green lines are results of data fittings for the Ti⁴⁺ and Ti³⁺ ions, respectively. (b) Ti³⁺/(Ti³⁺ + Ti⁴⁺) ion ratio at an emission angle of 0° (blue symbols) and carrier density deduced from the Hall effect (red symbols) as functions of irradiation time. (c) Angle dependence of the ratio of Ti³⁺/(Ti³⁺ + Ti⁴⁺) obtained at different irradiation times. Solid lines are results of data fittings based on Eq. (1).

the oxygen-deficient layer is almost unchanged though the percentage of the Ti^{3+} species keeps growing with irradiation time.

Provided that the V_os donate mobile electrons, the oxygen deficient surface layer will be conducting when the content of V_os is high enough. As shown in Fig. 3, ion irradiation indeed causes a resistive transition: After Ar⁺ irradiation, the originally insulated film becomes metallic above 100 K and semiconducting below ~100 K. Figures 3(a) and 3(b) show the sheet resistance (R_S) of the TiO₂ film as a function of temperature. R_S displays a linear variation against T² in the temperature range of 100 K < T < 300 K. This kind of temperature dependence is a typical character of the Fermi liquid systems, arising from electron-electron scattering.

In the low temperature range, an alternative mechanism dominates the transport behaviour. As also shown in Figs. 3(a) and 3(b), when the sample is cooled across 100 K, a resistance upturn emerges, leaving behind a local minimum at ~92 K. A close inspection reveals distinct temperature dependences of sheet resistance below 100 K. As shown in Fig. 3(b), R_S of the film irradiated for 0.5 min displays a linear variation against $1/T^{1/3}$. The magnitude of the R_S upturn thus resulted is very large. From 100 K down to 2 K, the sheet resistance grows from 9500 to 39700 Ω/\Box , fourfold enhanced. This change does not obey an exponential growth upon cooling as expected for a strong localization system but it is well beyond the scope of weak localization. Possibly, this sample locates at the border between the weak and strong localization. Indeed, strong localization may have taken place at 2 K since $R_{S}(2 \text{ K}) = 39.7 \Omega/\Box$ exceeds the quantum of the resistance $h/e^2 = 25.8 k\Omega$ but does not at 100 K where $R_{S}(100 \text{ K}) = 9.5 \text{ k}\Omega/\Box$, here h is the Planck constant. At present, the exact mechanism that governs the $R_S \propto 1/T^{1/3}$ dependence is still not very clear. However, we noted a theoretical prediction of a T^{1/3}-term correction to one-dimensional conduction, arising from electron-electron



FIG. 3. (a) and (b) Temperature dependence of sheet resistance of the films irradiated for 0.5, 1, 2, and 3 min, presented in different temperature scales. Solid lines are guides to the eye, showing a T^2 dependence of R_S in (a) and a lnT or a $1/T^{1/3}$ dependence in (b). (c) and (d) Carrier density (n_S) and Hall mobility (μ_H) of the films irradiated for 0.5, 1, 2, and 3 min.

collisions with small energy transfer.²⁴ There are also reports about the $1/T^{1/3}$ -power-law growth of the resistance for onedimensional nanowires,^{25–27} though the magnitude of the resistance upturn there (~20%) is much lower than that observed here. Presumably, at low temperatures, the network of conduction paths in the 0.5-min-irradiated TiO₂ film is quasi-one-dimensional in nature.

For the samples irradiated for a time longer than 1 min, however, a linear relation between R_S and ln(1/T) appears. This is the typical behaviour of a two-dimensional system in weak localization or a system suffering from Kondo scattering. Supported by the investigation of the negative MR shown later, we prefer to ascribe the resistance upturn upon cooling to the Kondo effect. As for the R_S of the 1min-irradiated film, it is in an intermediate state between the one- and two-dimensional conductions. As a result, its temperature dependence is neither $1/T^{1/3}$ nor ln(1/T).

As shown in Fig. 3(c), ion irradiation indeed generates mobile electrons. The carrier density is $n_{\rm S} \approx 5 \times 10^{13} \, {\rm cm}^{-2}$ after the 0.5-min irradiation, nearly independent of temperature except for a slight decrease below 30 K. Increasing irradiation time leads to a monotonic growth in carrier density. The maximal n_s is $\sim 1.0 \times 10^{14}$ cm⁻², gained at 30 K in the sample irradiated for 3 min. In Fig. 2(b), we show the carrier density as a function of irradiation time, obtained at roomtemperature, and observed an exact correspondence between carrier density and the content of Ti³⁺ ions. This correspondence implies that the mobile electrons exclusively come from Ti^{3+} ions. In Fig. 3(d), although with the decrease in temperature, $\mu_{\rm H}$ displays a sizable increase from $10 \,{\rm cm}^2/{\rm V}$ s at room temperature to 19 cm²/V s at 100 K for the 1- and 2-min-irradiated films, and the hall mobility decreases below 100 K, concomitant with the occurrence of the resistance upturn [Fig. 3(b)].

To get a further insight into the mechanism that determines the electronic transport process in our 2DEG, the MR is studied in perpendicular magnetic fields. As shown in Fig. 4(a), the 2DEG generated by the 0.5-min irradiation displays a positive MR = $R_S(H)/R_S(0) - 1$ at low temperatures. Surprisingly, the MR ratio is unusually large, exceeding 16% under an applied field of 7 T at 2 K. It cannot be ascribed to the classical orbital effect that will produce a MR around 0.0001% when carrier mobility is 4 cm²/V s as shown in Fig. 3(d). In fact, shrinkage of electron wave function in the magnetic field could be responsible for the positive MR, which is particularly strong when the electron wave functions are centred on oxygen vacancies but they still have finite overlap.²⁸

Increasing irradiation time to 1 min, the MR-B relation becomes much more complex. With the increase in the magnetic field, the MR at 2 K experiences first a finite decrease from 0 to a finite negative value and then a rapid increase to values well above zero. These two processes are also observed in the MR-B curve recorded at 5 K, though the second process only produces a weak resistance upturn. The sample irradiated for a time longer than 1 min exhibits negative MR in the whole temperature range investigated. As shown in Figs. 4(c) and 4(d), the MR-B curves are all bellshaped, and the MR magnitude becomes more prominent with the decrease in temperature. The maximal MR, under



FIG. 4. Out-of-plane magnetoresistance of the TiO_2 films irradiated for different times. The symbols and solid curves represent the experimental and calculating results, respectively. For clarify, the data of 2 K and 5 K correspond to the left coordinate while others are the right.

an applied field of 7 T, is -2.5% at 2 K, appearing in the 3-min-irradiated sample.

 Ar^+ irradiation yields oxygen vacancies, introducing magnetic moments (1 μ_B per Ti³⁺) into the 2DEG. The presence of a considerable amount of Ti³⁺ ions has been confirmed by XPS analysis. In general, the local moments will interact with conduction spins, yielding an additional electron scattering. The improvement of the alignment of the local moments in an applied field will produce a negative MR that can be described by an equation obtained by Khosla and Fischer²⁹ based on the hypothesis of MR $\propto M^2$ (M is magnetization)

$$MR = -a^{2}\ln(1 + b^{2}B^{2}) + c^{2}B^{2}/(1 + d^{2}B^{2}).$$
(2)

The first term of Eq. (2) describes the negative MR whereas the second term is the ordinary MR arising from the classical orbital effect when two species of charge carriers coexist. As shown by the solid curves in Fig. 4, Eq. (2) well reproduces the experimental results for the 2- and 3-min-irradiated 2DEGs and the negative part of the MR for the 0.5- and 1-min-irradiated 2DEGs in the whole field range. The fitting parameter b is proportional to 1/T at high temperatures (Fig. 5), in good agreement with the prediction of Khosla and Fischer. The deduced b-1/T slope is 5.2 K/T above 20 K for the 3-min-irradiated sample. These behaviors are very similar to those of the diluted magnetic semiconductor which exhibits a b-1/T slope of 5.0 K/T for oxygen deficient ZnO³⁰ and 8.6 K/T for Nb-doped TiO₂.³¹ Ferromagnetism has been observed in experiments and analysed via first-principles calculations in systems of undoped HfO₂, TiO₂, In₂O₃, and ZnO,^{32–35} and general consensus attributes it to the spinpolarized lattice defects. These results suggest that the negative MR stems mainly from a coherent magnetic scattering of the itinerant electrons by correlated localized moments.



FIG. 5. Fitting parameter b in Eq. (2), as a function of reciprocal temperature.

We also tried to fit the MR-B relation to the model of weak localization, and the results are not satisfactory especially under high applied fields.

In summary, high quality anatase TiO2 thin films were fabricated on LAO substrates. The 2DEG with a width of 4 nm is achieved by introducing oxygen vacancies into the surface layer of TiO₂ via argon-ion irradiation. Distinct transport behaviours are observed, characterized by a T-squared dependence of the sheet resistance above $100 \,\mathrm{K}$ or a $\mathrm{T}^{-1/3}$ or ln(1/T), depending on irradiation time, dependence below 100 K. For the 2DEG produced by short time irradiation and thus with a low carrier density, unusual large positive magnetoresistance that cannot be explained by the classical orbital effect is observed in the perpendicular magnetic field. With the increase in carrier density, the magnetoresistance changes from positive to negative can be ascribed to the depression of magnetic scattering. The present work demonstrates the possibility of gaining high quality TiO2-based 2DEG based on ion irradiation that offers opportunities for the fabrication of multifunctional nanometer-sized materials and devices.

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