

Realization of a High-mobility Two-dimensional Hole Gas at a SrTiO₃ Interface and Control of the Carrier Type

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Abstract

Despite intensive studies on two-dimensional *electron* gas (2DEG) formed at the SrTiO₃ (STO) interface [1], forming a 2D *hole* gas (2DHG) at the STO interface is extremely difficult [2], although both are essential for the realization of high-speed oxide-based electronics. Here, we demonstrate a very simple method to realize a 2DHG with an ultrahigh mobility of 24,000 cm²V⁻¹s⁻¹ at an STO interface [3]. The 2DHG is obtained by depositing a sub-nm-thick Fe layer (thickness $t \leq 0.2$ nm) on STO substrates at room temperature in an ultrahigh vacuum chamber. The Fe layer is oxidized and becomes amorphous FeO_y ($y \approx 1.5$). Magnetotransport measurements reveal the existence of high-mobility carriers in the STO side, and the carrier type changes from a *pure p*-type ($t \leq 0.2$ nm) to *n*-type ($t > 0.3$ nm) by varying the Fe thickness. In a *p*-type sample ($t = 0.1$ nm), the Shubnikov - de Haas oscillation is clearly observed by applying a magnetic field perpendicular to the film plane but disappears with an in-plane field. These results clearly demonstrate the 2D nature of the high-mobility hole carriers.

1. Introduction

The two-dimensional hole gas (2DHG) at the strontium titanate (STO) interface has long been pursued since the discovery [1] of its counterpart—the 2D electron gas (2DEG)—and various related emergent physical phenomena [4-9]. This 2DHG is expected to be rich of unexplored hole-related physics. The combination of a 2DEG and 2DHG at an STO surface is also promising for oxide-based electronic devices, such as ultra-violet light-emitting diodes, high-mobility diodes and transistors. In principle, the 2DHG is expected to form at the interface between the non-polar SrO layer of STO and a negatively charged atomic layer, such as the AlO₂ layer of LaAlO₃. However, this interface mostly turned out to be insulating [1] in previous experimental tests. In the only successful observation of the 2DHG thus far [2], the hole mobility was relatively low (1000 cm²/Vs), and a meticulous growth-and-annealing process was inevitably required to produce a sharp interface without oxygen vacancies. In addition to these difficulties, the characterization of this 2DHG was not straightforward due to the parallel conduction of the co-

existing 2DEG. A completely isolated and well-controlled 2DHG at the STO interface is thus required to access the unexplored physics and capabilities of this system.

2. Experimental results

We prepare the 2DHG samples using TiO₂-terminated STO (001) substrates via molecular beam epitaxy (MBE). After introducing STO substrates in an ultrahigh vacuum MBE chamber with a background pressure of 3×10^{-8} Pa, a very thin Fe layer with a nominal thickness t_{Fe} (nm) is deposited with a slow growth rate of 0.03 nm/min at a substrate temperature of 50°C. Finally, the samples are capped with 1-nm-thick Al at the same temperature (Fig. 1). We examine a series of samples A10, A15, A20, A25, A30, A40 and A60 with various thicknesses $t_{\text{Fe}} = 0.10, 0.15, 0.20, 0.25, 0.30, 0.40,$ and 0.60 nm, respectively. The Fe and Al layers are completely oxidized after being taken out of the MBE chamber and transform into AlO_xFeO_y layers. Using atomic force microscopy (AFM) and high-resolution scanning transmission electron microscopy (STEM), we confirm an atomically flat STO interface and a nearly amorphous structure of the AlO_x/FeO_y top layers.

By measuring the Hall effect, we find that the samples with $t_{\text{Fe}} < 0.25$ nm show *p*-type conduction and that the carrier type is switched to *n*-type when $t_{\text{Fe}} > 0.25$ nm. Only in sample A25 with an intermediate t_{Fe} value (= 0.25 nm), the magnetic field B dependence of the Hall resistances R_{yx} is strongly nonlinear, showing both negative and positive B -dependences, which implies the coexistence of both holes and electrons. Meanwhile, in sample A10 with the smallest value of t_{Fe} (= 0.1 nm), the $R_{yx} - B$ curve has a positive slope and is linear up to 55 T, indicating pure *p*-type conduction. To date, *p*-type conduction has never been reported when depositing a metallic element on the surface of STO substrates. Samples from A10 to A40 show similar metallic behavior; the sheet resistance R_{sheet} decreases by two orders of magnitude upon cooling and is saturated below 10 K. These features, such as the anomalous carrier type change with the layer thickness and strong dependence of the resistivity on temperature T , are clearly distinct from the transport properties of Fe or impurity-doped FeO_y thin films. The hole mobility in sample A10 reaches 24,000 cm² V⁻¹ s⁻¹ below 10 K (Fig. 1), which is the highest value ever reported for *p*-type oxides and comparable to the best values of the 2DEG.

We also conduct Seebeck effect measurements to verify the carrier type and its dependence on t_{Fe} . In sample A20, a positive Seebeck coefficient $S = 150 \mu\text{V K}^{-1}$ is obtained, indicating p -type conduction. Meanwhile, in sample A40, the negative $S = -160 \mu\text{V K}^{-1}$ indicates n -type conduction. These Seebeck coefficients are in the same magnitude ($\sim 150 \mu\text{V K}^{-1}$) as those typically reported for bulk STO or STO-based heterostructures [10]. These results are consistent with the Hall effect results mentioned above, confirming that both p -type and n -type conduction occur at the STO interface.

Figure 2 shows the Shubnikov - de Haas (SdH) oscillations measured in sample A10. At 500 mK, the longitudinal resistance R_{xx} shows strong oscillations when we apply a magnetic field B perpendicular to the film plane, as shown in the second derivative characteristics ($d^2R_{xx}/dB^2 - 1/B$ curves) with $B // [001]$ in Fig. 2. From Fourier transform (FT) analysis of the $d^2R_{xx}/dB^2 - 1/B$ curves, we see that there are two components of the oscillation; one component has a low frequency $f_1 = 38.9 \text{ T}$, corresponding to the weak oscillation at low B (see “•”), and the other component has a high frequency $f_2 = 116.29 \text{ T}$, corresponding to the strong and high-frequency oscillation at high B (see “▽”). These oscillations correspond to two kinds of hole carriers with different effective masses (m_1^* , m_2^*) and densities (p_1 , p_2): The light carrier corresponds to the component f_1 with $m_1^* = (1.43 \pm 0.17) m_0$ (m_0 is the mass of a free electron) and $p_1 = 1.89 \times 10^{12} \text{ cm}^{-2}$, and the heavy carrier corresponds to the component f_2 with $m_2^* = (2.57 \pm 0.15) m_0$ and $p_2 = 5.66 \times 10^{12} \text{ cm}^{-2}$. Both the f_1 and f_2 oscillatory components disappear when we apply B in the in-plane direction. This result indicates that both light and heavy holes are 2D carriers confined at the STO interface.

3. Conclusions

We reported for the first time that one can create a completely isolated 2DHG with an extremely high mobility up to $\sim 24,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ by depositing an angstrom-thin Fe layer on top of an STO substrate nearly at room temperature ($\sim 50^\circ\text{C}$). This is the highest hole mobility ever reported for oxides. Furthermore, we demonstrate the ability to control the carrier type from p -type (2DHG) to n -type (2DEG) by increasing the Fe thickness or by thermal annealing. This new and easy method of forming both 2DHG and 2DEG at an STO interface will facilitate the development of high-speed electronic devices and circuits on STO interfaces at an extremely low cost.

Acknowledgements

This work was partly supported by Grants-in-Aid for Scientific Research (No. 18H03860, 17H04922), the CREST Program (JPMJCR1777) of the Japan Science and Technology Agency, and the Spintronics Research Network of Japan (Spin-RNJ). A part of this work was conducted at Advanced Characterization Nanotechnology Platform of the University of Tokyo, supported by “Nanotechnology Platform” of the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan, and the Cryogenic Research Center, the University of Tokyo.

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Figures

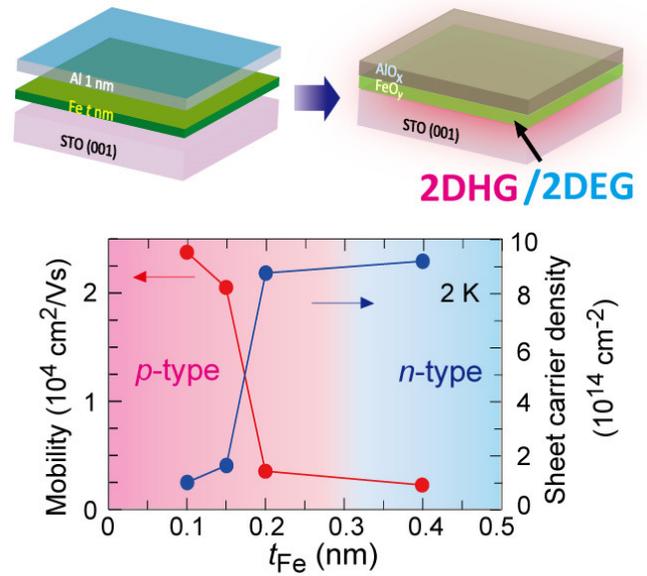


Fig. 1. Deposited layered structure in the MBE chamber (left) and actual structure of the studied samples after taken out of the MBE chamber (right). Mobility and sheet carrier density as functions of Fe thickness (Bottom).

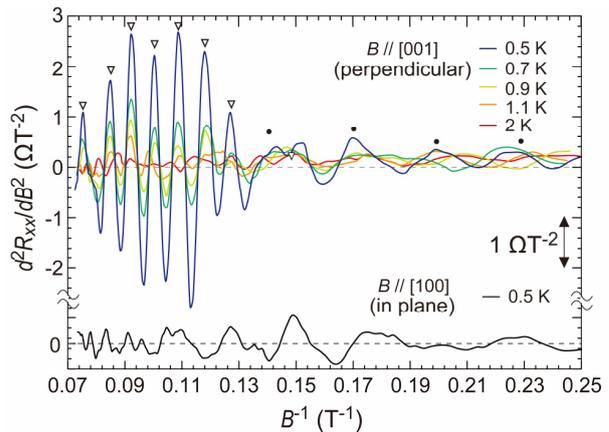


Fig. 2. $d^2R_{xx}/dB^2 - 1/B$ curves measured at various temperatures with $B // [001]$ (top) and at 500 mK with $B // [100]$ (bottom). There are two oscillatory components when B is applied perpendicular to the film plane (parallel to the $[001]$ direction), which are marked by black inverted triangles and circles.