Tuning of metal-insulator transition of two-dimensional electrons at parylene/SrTiO$_3$ interface by electric field

H. Nakamura,1,2 H. Tomita,1,2 H. Akimoto,3 R. Matsumura,1 I. H. Inoue,5 T. Hasegawa,2 K. Kono,3 Y. Tokura,2,4 and H. Takagi1,2,3

1Department of Advanced Materials, University of Tokyo, Kashiwa 277-8561, Japan.
2Correlated Electron Research Center (CERC), National Institute of Advanced Industrial Science and Technology (AIST), AIST Tsukuba Central 4, Tsukuba 305-8562, Japan
3Advanced Science Institute, RIKEN (The Institute of Physical and Chemical Research), Wako 351-0198, Japan
4Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan

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Electrostatic carrier doping using a field-effect-transistor structure is an intriguing approach to explore electronic phases by critical control of carrier concentration [1, 2]. We demonstrate the reversible control of the insulator-metal transition (IMT) in a two dimensional (2D) electron gas at the interface of insulating SrTiO$_3$ single crystals. Superconductivity was observed in a limited number of devices doped far beyond the IMT, which may imply the presence of 2D metal-superconductor transition. This realization of a two-dimensional metallic state on the most widely-used perovskite oxide is the best manifestation of the potential of oxide electronics.

The perovskite SrTiO$_3$ is sometimes called the “silicon of oxide electronics” because it is commonly used as a substrate for epitaxial growth of a variety of oxide films [3]. Not only it is an insulator with a band gap of 3.2 eV but also a quantum paraelectric with an extremely large dielectric constant of more than 10$^5$ at low temperatures [4]. N-type conduction has been realized by introducing a small number of oxygen vacancies or by cation substitutions. An insulator to metal transition occurs at a rather low carrier concentration of $n \sim 10^{18}$ cm$^{-3}$ [4, 5] for a doped oxide system, which owes largely to the high mobility exceeding 10$^3$ cm$^2$/Vs. Superconductivity emerges in a limited concentration range from $n = 7 \times 10^{18}$ to $5 \times 10^{20}$ cm$^{-3}$ and the transition temperature $T_c$ shows a bell shaped dependence on $n$ with an optimum $T_c \sim 0.35$ K at $n \sim 10^{20}$ cm$^{-3}$ [6]. The carrier concentration required to achieve superconductivity is two orders of magnitude smaller than that for high-$T_c$ cuprates which is a great advantage to realize electrostatic control of insulator-metal and superconductor transitions.

Such a unique doping properties of SrTiO$_3$ has triggered attempts to dope charged carriers electrostatically by using a field effect transistor (FET) structure. However, so far, attempts to realize metallic state have been unsuccessful [7]. This is due to several reasons, notably carrier trapping at the gate insulator/SrTiO$_3$ interface, Schottky barrier formation between SrTiO$_3$ and source and drain electrodes, and insufficient breakdown voltage of the gate insulators. An alternative approach has shown that two dimensional electron gas can be created at SrTiO$_3$/LaAlO$_3$ hetero-interface by a charge-transfer due to a polar discontinuity [8] or possible oxygen defects [9, 10] or the electronically tailored interface was found to be superconducting as in the bulk material [11, 12].

Recently the performance of SrTiO$_3$ FET has been drastically improved [13, 14] by overcoming the contacts and interface problems. These new devices showed metallic behavior down to 7 K [15]. In this Letter, we extend our study to lower temperatures below 1 K, and show that the two-dimensional insulator-metal transition is indeed achieved at the interface between parylene and SrTiO$_3$ reversibly while sweeping the gate voltage. A superconducting transition to zero-resistance state was observed at 0.13 K only in a sample where a large enough number of carriers were successfully introduced beyond the critical carrier number for the insulator-metal transition (IMT). The observation of two dimensional insulator-metal-superconductor transition represents major progress not only in the attempts for future oxide electronics but also in oxide physics.

A top gate FET was fabricated consisting of Au/parylene/SrTiO$_3$ layers. Al electrodes of the Hall bar geometry (Fig.1a) were evaporated on an atomically flat surface of SrTiO$_3$ (100) before depositing the gate insulator parylene. The use of Al electrode is the key to suppressing Schottky barrier formation. Details of the device fabrication are described elsewhere [16]. We have measured six samples (Sample A-F) with different parylene thickness (0.53-0.63 µm), corresponding to capacitance $C_i = 4.4 - 5.3$ nF/cm$^2$ allowing a wide-ranging threshold gate voltage ($V_{th} = 97-170$ V). Resistivity measurements above 2 K were carried out by dc four-probe method with temperature control by the Quantum Design Physical Property Measurement System, while those below 1 K were carried out by ac four-probe method (frequency 45 Hz) in a dilution refrigerator.

Figure 1a shows the temperature dependence of sheet resistance for various gate voltages (or electron densities). As the gate voltage is increased, the temperature dependence changes from insulating to metallic. At high enough gate voltages, the resistance eventually becomes metallic all the way down to a low tempera-
The above behavior suggests the emergence of 2D metal at the parylene/SrTiO$_3$ interface. This is in contrast to the scaling theory of localization, which predicts that a 2D system should become insulating at zero temperature [20]. Recent experiments on very high mobility 2D electron gases formed in Si and GaAs hetero-structure, however, demonstrated the existence of insulator-metal transitions even in 2D and have been attracting considerable attention [21]. To explore the ground state of electrostatically induced metallic inter-

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two independent mobilities are compared; the field-effect mobility \( \mu_{\text{FE}} = (1/e)(\sigma_{2D}/dn_{2D}) \), where \( \sigma_{2D} = 1/R_{2D}, \) \( n_{2D} = C_2V_G/e, \) and the magnetoresistance mobility \( \mu_{\text{MR}} \) estimated by the magnitude of the quadratic part of magnetoresistance \( \Delta R/R_0 = (\mu_{\text{MR}}B)^2 \). While the two mobilities agree very well in the larger gate voltage region above the kink, they are distinct from each other in the transition region (Fig. 3b). The \( \mu_{\text{MR}} \) is as large as 2000 \( \text{cm}^2/\text{V}s \) and surprisingly independent of carrier density above and below the kink. In contrast, \( \mu_{\text{FE}} \) shows a rapid decrease with decreasing voltage around the transition region. This contrasting behavior between the two mobilities may be understood naturally in terms of phase separation. The conduction should be dominated by the metallic domains. The magnetoresistance is then almost identical to those of metallic domains independent of the gate voltage. On the other hand, the value of \( \mu_{\text{FE}} \) reflects the channel resistance. Narrowing of the metallic domain during the separation enhances the channel resistance and hence \( \mu_{\text{FE}} \) decreases rapidly. Thus, the transition between metallic and insulating state tuned by a gate voltage is very likely a first order transition accompanied by a phase separation. The origin of seemingly weak localization behavior in the transition region shown in Fig. 1 is not clear. We suspect that narrow percolation path in the mixed phase might be responsible for the apparent logarithmic temperature dependence.

The discontinuous transition presented here is reminiscent of the “steplike” change in conductance observed in SrTiO\(_3\)/LaAlO\(_3\) hetero-structures as a function of LaAlO\(_3\) thickness \[14\], where the electric field produced by polar discontinuity is believed to play a role. In both cases, the sheet resistance of the conducting side of the “discontinuous change” is of order 10 k\( \Omega \)m, suggesting there exists a universal minimum metallic conductivity of around \( \sim 10^{-4} \text{S} \). Although the mechanism of first-order-like metal-insulator transition is currently unclear, the observation of similar behavior in different types of device structure seems to indicate that the origin of discontinuous metalization may be intrinsic at least to SrTiO\(_3\).

In only a few samples, we were able to apply a large enough gate voltage without breaking the gate insulator (parafilm) to go far beyond the discontinuous IMT. With successful accumulation of an enough number of carriers, we observed zero-resistance state associated with superconductivity. As shown in Fig. 4, at 128 mK the resistance decreases gradually with increasing gate voltage, namely the carrier concentration, and eventually becomes zero within our resolution above \( n_{2D} \sim 2 \times 10^{12} \text{cm}^{-2} \). With decreasing gate voltage, then, the resistance goes back to a finite value reproducibly. With increasing temperature from 128 mK at \( n_{2D} \sim 2 \times 10^{12} \text{cm}^{-2} \), the resistive transition to a normal state is observed around 130-160 mK as seen in the inset of Fig. 4. The application of magnetic field of 0.03 T perpendicular to the plane was found to suppress the zero resistance state.

These observations demonstrate that superconductivity is marginally achieved around \( n_{2D} \sim 2 \times 10^{12} \text{cm}^{-2} \).
FIG. 4: Continuous and reversible electrostatic tuning of superconductivity at the surface of undoped SrTiO$_3$ single crystal. Reversible and continuous control between the zero-resistance and the finite resistance was achieved by sweeping the gate voltage at $T = 128$ mK. The inset shows temperature dependence of the sheet resistance at a constant sheet carrier density at $B=0$ T (continuous line) and at $B=0.03$ T (dashed line). Zero-resistance is seen below 130 mK for $B=0$ T.

This sheet carrier density corresponds to $n_{3D} \approx 2 \times 10^{18}$ cm$^{-3}$ assuming the thickness of 2D electron layer to be 10 nm. Though the reason is not obvious, this is similar to the onset $n_{3D} \approx 7 \times 10^{18}$ cm$^{-3}$ reported for bulk, given the uncertainty in 2D electron layer thickness. Note that we did not see any trace of superconductivity in the metallic sample shown in Fig. 1c, down to 15 mK at $n_{2D} \sim 1 \times 10^{12}$ cm$^{-2}$. These suggest that the 2D insulator-metal transition occurs first and that, to achieve 2D metal-superconductor transition, extra charge carrier must be introduced. We, however, cannot rule out completely superconductivity at an extremely low temperature in the sample near IMT.

It should be noted that by applying a large gate field for a long period (typically $V_G > 150$ V for one week), another superconductor-like transition without a zero resistance state appears around 0.35 K which does not disappear even after reducing the gate field. This is highly likely to be the superconductivity associated with the creation and/or migration of oxygen defects at the interface.

In conclusion, we have demonstrated that the parylene/SrTiO$_3$ interface shows a transition from an insulator into two-dimensional metal by the electrostatic carrier doping as observed in high mobility 2D electron gas formed in Si and GaAs hetero-structures. We believe that this represents a breakthrough in oxide electronics. One of the unique features of the parylene/SrTiO$_3$ interface is a zero-resistance state emerging at 130 mK. The emergent superconductivity is thought-provoking not only because of the possible 2D character but also because it develops in a electron gas with a small Fermi energy of a few meV—usually trivial interactions like spin-orbit coupling [27] can be comparable with Fermi energy in this system and may give rise to an exotic superconductivity. Therefore, what we have demonstrated in this study is not simply a hectic chase of oxide electronics after the conventional semiconductor but also a prelude to new paradigm of transition metal oxide physics.

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