Electric-field control of Mott transition in electrolyte-gated (Nd,Sm)NiO₃ thin film

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1. Introduction

Development of information and communication technologies relays on the miniaturization of device scales leading to the improvement of its performance. However, the miniaturization of device scales is expected to face physical and technological limits. In order to overcome this issue, new devices based on new materials and different principles of physics, so-called beyond CMOS technologies, have been intensively studied. One of promising candidates of beyond CMOS technologies is a field effect transistor (FET) based on a metal-insulator transition (MI) in correlated electron materials, so-called Mott transistor. Mott transition (a MI transition caused by the interplay of electron correlations and kinetic energy) can be controlled by carrier doping.[3] However, it is hard to induce the Mott transition in correlated electron materials by using a conventional FET structure, because the maximum capacitance of the conventional gate dielectrics such as SiO_2 (~ 10^{13} cm⁻²) is never enough to induce the Mot transition. For example, sheet carrier density about 10¹⁴ to 10¹⁵ cm⁻² is necessary to be doped in order to induce the Mott transition in a high- $T_{\rm C}$ cuprate or a colossal magnetoresistance (CMR) manganite.[4] These values are more than 10 times larger than those achievable using conventional gate dielectrics. Therefore, for the development of Mott transistor, correlated-electron materials with a MI transition attainable at significantly lower carrier concentrations are required

In this study, to demonstrate an electric-field control of the Mott transition, we used rare-earth nickelates RNiO₃ (R = Nd, Sm) for the channel material. RNiO₃ displays a temperature-driven sharp MI transition due to the strong electron correlations and the transition temperature $(T_{\rm MI})$ can be reduced by a chemical carrier doping. For example, in $Nd_{1-x}Ca_xNiO_3$, T_{MI} decreases with increasing x. Here, the substitution of Ca2+ for Nd3+ provides a hole. The ratio $dT_{\rm MI}/dx$ is approximately -3200 K/hole.[5] From this value, we can estimate that the accumulation of holes with a sheet carrier density of $\sim 10^{14}$ cm⁻² can reduce $T_{\rm MI}$ of $NdNiO_3$ by about 100 K. The reduction of T_{MI} enables us to obtain a large resistance change just below $T_{\rm MI}$. However, the sheet carrier density of ~10¹⁴ cm⁻² is still too high to achieve by electrostatic methods using ordinary gate dielectrics. In response, we have employed an ionic liquid

electrolyte as the gate dielectric. An electric double layer (EDL) at the interface between the ionic liquid and the channel material can reach a sheet carrier density of 10¹⁵ cm⁻². Using the EDL transistor (EDLT) structure, we have succeeded in the electric-field control of a MI transition in (Nd,Sm)NiO₃ channels at room temperature.

2. Experiments

 $Nd_{1-x}Sm_xNiO_3$ [NSNO(x)] thin films were deposited on $NdGaO_3$ (110) substrates using the pulsed laser deposition technique. X-ray diffraction (XRD) measurements showed that the single-phase NSNO(x) films were epitaxially grown on the substrates. The thickness of the NSMO(x) films was estimated from Laue fringe peaks observed near the (220) diffraction peak.

The NSNO(x) film was patterned with a channel area of $10 \times 100~\mu\text{m}^2$ using the photolithography and Ar-ion milling techniques. Layered Au/Pt electrodes for gates, sources, drains, and voltage probes were deposited by electron beam evaporation. The optical view of our EDLT is shown in Fig. 1

Both the gate electrode and the channel area were covered by the ionic liquid N,N-diethyl-N-(2-methoxyethyl)-N-methylammonium tetra fluoroborate (DEME-BF₄). The nominal capacitance C of our EDLT was about $10~\mu F/cm^2$. From this value, we can estimate a sheet carrier density of $\sim 1.5 \times 10^{14}~cm^{-2}$ for $V_G = -2.5~V$ which is enough to control the $T_{\rm MI}$ of NSNO channel. To avoid the effect of contact resistances, we measured the channel resistivity ρ by a standard four-probe configuration. The temperature T dependence of ρ of the electrolyte-gated NSNO(x) thin film was measured from 300 to 20 K at a cooling/heating rate of 3 K/min. The gate voltage dependence of drain current I_D was measured just below the $T_{\rm MI}$ of NSNO(x) channel.

3. Results and discussions

Figure 2(a) shows ρ -T curves of EDLT with NSNO(0.5) channel at gate voltage $V_{\rm G}=0$ and -2.5 V. By applying a gate voltage, the $T_{\rm MI}$ is shifted from 288 K to 256 K. This result indicates that as expected, the hole-carrier accumulation reduce the $T_{\rm MI}$ of NSNO(0.5) channel at room temperature. The resistivity ρ below the $T_{\rm MI}$ for $V_{\rm G}=0$ V changes by more than one order of magnitude.

Figure 2(b) shows the $V_{\rm G}$ dependence of $I_{\rm D}$ in the NSNO(0.5) channel at 280 K. The $I_{\rm D}$ is increased by about 400 % when the $V_{\rm G}$ is decreased below -1.5 V. A large hysteresis of the $I_{\rm D}$ can be expected to result from the slow movement of ions in the ionic liquid in response to the gate voltage. Notably, our EDLTs show a high effective mobility $\mu_{\rm eff}$. From Fig. 2(b), $\mu_{\rm eff}$ of the NSNO(0.5) channel can be estimated to be ~740 cm²V⁻¹s⁻¹ for $V_{\rm G}$ = -2.2 V. Such a large $\mu_{\rm eff}$ is due to the strong nonlinear dependence of the mobile carrier density on doping - characteristic of filling-controlled Mott transitions.

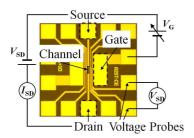


Fig. 1. Optical view of EDLT with the measurement configuration.

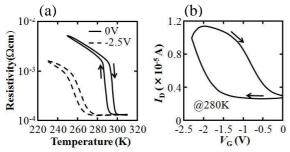


Fig. 2 (a) Resistivity – temperature curves of the EDLT with NSNO(0.5) channel at gate voltage $V_G = 0$ and -2.5 V. (b) V_G dependence of I_D of the NSNO(0.5) channel at 280 K.

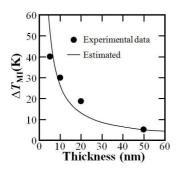


Fig. 3 The channel thickness d dependence of $\Delta T_{\rm MI}$ for NdNiO₃ EDLTs. Solid line represents the d dependence of $\Delta T_{\rm MI}$ estimated from the capacitance of the EDLT and the $dT_{\rm MI}/dx$ of NdNiO₃.

Figure 3 shows the channel thickness d dependence of $\Delta T_{\rm MI}$ for NdNiO₃ EDLTs, where $\Delta T_{\rm MI} = T_{\rm MI} (V_{\rm G} = -2.5 \text{ V})$ -

 $T_{\rm MI}(V_{\rm G}=0)$. The $\Delta T_{\rm MI}$ decreases with increasing the d. The solid line represents the d dependence of $\Delta T_{\rm MI}$ estimated from the $C = 10 \mu F/cm^2$ of our EDLT and the $dT_{MI}/dx =$ -3200 K/hole of NdNiO₃ and by assuming the accumulated carriers distribute homogeneously in the channels along the depth direction. As seen in Fig. 3, the experimental data fit well with the solid line, suggesting the homogeneous distribution of accumulated carriers. This result seems to contradict the anticipated electrostatic carrier accumulation, because the thickness of the carrier-accumulation layer must be within the Thomas-Fermi screening length, which does not depend on the d. Although the mechanism of the homogenous distribution of accumulated carriers has not been understood, we think that this phenomenon is one of the peculiarities of the electric-field effect on correlated electron materials.

4. Conclusions

We have demonstrated the electric-field control of the MI transitions in (Nd,Sm)NiO₃ channels by accumulating hole carriers with EDL technique. We found the two characteristics of the electric-field effect on correlated electron materials; an apparently high mobility and a homogeneous distribution of accumulated carriers. We believe that this study can be potentially applicable to Mott transistor and calls for further understanding of the mechanism in the electric-field control of phase transitions in correlated electron materials.

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