

Metal-insulator Transition Field Effect Transistor Observed by 4-probe Lamination Contact Electrode

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Abstract

Gate electric field induced metal-insulator transition on the charge order phase in α -(BEDT-TTF)₂I₃ single crystal field effect transistor (FET) was clearly observed just below charge order phase transition temperature. Obvious increase of the electrical conductance induced by the applied gate electric field and its temperature dependence was observed, which corresponds to partial dissolution of the charge-order phase by the effect of injected carrier.

1. Introduction

Strongly correlated electron materials and their various electronic phase such as Mott insulator, charge order, and so on, attracted wide interest in material science field. However, in spite of the extensive interest in strongly correlated organic materials and the effect of a gate electric field on the field-effect transistor (FET) structure[1], only a few studies on the effect of a gate electric field have been conducted[2,3], which is mainly due to the technical difficulties with sample preparation. Samples to observe the gate electric field effect must possess at least the three following conditions: (1) a mirror crystal surface, (2) an inert interface between the crystal surface and the gate insulator, and (3) a sufficiently thin crystal for adherence onto the substrate and for distinguishing enhanced electrical current by the gate electric field from the bulk electrical current.

We introduced novel technique on measurement of single crystal organic field effect transistor. Lamination contact electrode (LCE) in this study is thin film Au 4-probe electrode pattern that is formed on the thin parylene film. LCE was so thin that LCE spontaneously stick on the surface of organic single crystal very softly, and can establish electrical contact between Au thin film electrode pattern and crystal surface without any chemical or thermal damage. In the early stage of the research works on organic single crystal devices, thin organic crystal was grown and laminated on the SiO₂/Si substrate surface[4]. However, this method was only effective on very thin platelet organic crystal because bulky crystals cannot stick on SiO₂ surface. On the other hand, LCE exchanged the role of organic single crystal and substrate, therefore, no restriction on the crystal side exist as far as normal 4-probe electrical measurement.

Figure 1(a) is the picture of the LCE during being peeled off from the preparation substrate. When the parylene thin

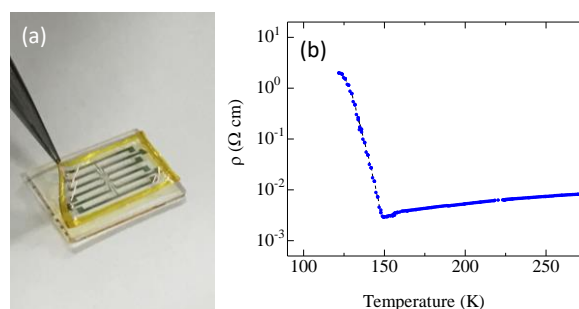


Fig.1 (a) Picture of the lamination contact electrode (LCE) being peeled off from the preparation substrate. When the parylene thin film is peeled off the preparation substrate, 4-probe Au electrode pattern simultaneously peeled off from the substrate. (b) Temperature dependence of electrical resistivity of α -(BEDT-TTF)₂I₃ measured by using 4-probe LCE. The metal insulator phase transition at around 140 K was clearly observed and almost no temperature hysteresis is observed, which demonstrates the reliability of the LCE.

film is peeled off the preparation substrate, Au electrode pattern fabricated below the parylene layer simultaneously peeled off from the substrate. The obtained LCE was laminated on the surface of organic single crystal.

Single crystals of α -(BEDT-TTF)₂I₃ were grown by electrocrystallization. BEDT-TTF powder and tetrabutylammonium iodide were dissolved in benzonitrile. A pair of Pt wire was used as the electrodes for electrocrystallization. These procedures were conducted in a N₂ glove box. The temperature of the growth cell was kept at 308 K during growth, and a constant electrical current of 0.4 μ A was applied during the growth. Grown crystals were handled in anhydrous ethanol and settled on a surface of polydimethylsiloxane (PDMS) coated Neoprim film. Thermal expansion coefficient of the Neoprim film was 58 ppm/K. Strain or slippage at the interface of the organic single crystal and substrate are effectively suppressed by using PDMS/Neoplen. Selected single crystals of α -(BEDT-TTF)₂I₃ on the substrate surface was electrically contacted by using 4-terminal LCE. Figure 1(b) is one example of temperature dependence of the electrical resistivity obtained by using LCE. Electrical contact by LCE is stable even in low temperature. Temperature hysteresis is also almost negligible in this condition. Fig. 2 is an optical micrograph of the thin α -(BEDT-TTF)₂I₃ single crystal after

sticking the 4-terminal lamination contact electrode on it. Thin single crystal of which thickness was approximately 1 μm was slightly seen below the lamination contact 4-terminal electrode and gate electrode.

Voltage terminal gap of the 4-terminal lamination contact electrode was 50 μm , and source-drain distance (channel length) and width of were 180 and 50 μm , respectively. LCE was placed on the single crystal with original $xyz\theta$ micro-stage. 4-terminal electrical measurements were conducted by applying drain voltage of -0.1 V. The electrical current measurement and drain voltage application was ADVANTEST R6245. 4-terminal voltage difference was measured by Keysight 3458A. The gate voltage was generated by a system source meter (Keithley 2635A)

Figure 3 shows results of the field effect measurements obtained just below metal-insulator transition temperature. Fig. 3 (a) shows transfer characteristics of the phase transition FET. G/G_0 is the relative conductance under application of gate voltage (V_G) normalized by the bulk electrical conductance under zero- V_G . G/G_0 begin to increase above 40 V of V_G and gradually increase with increasing positive gate voltage until 70 V and saturate up to 100 V. When V_G decrease, saturated value of $G/G_0 = 1.1$ is kept until 36 V and abruptly decreased to 1.0 below 24 V. In addition, G/G_0 begin to increase below -40 V in the negative gate voltage region and gradually increase with negative gate voltage until -55 V and then saturate up tp -100 V. Same as in the positive V_G region, in the decreasing V_G region, the saturated G/G_0 value was kept until -46 V of V_G and then abruptly decrease to 1.0 below -30 V. One could think that the observed G/G_0 is small from the viewpoint of the conventional FET, however, this due to the high bulk conductivity of the organic charge transfer complexes. Organic charge transfer complexes consist of molecular donor and acceptor which make carrier by electron transfer from donor to acceptor, therefore organic charge transfer complexes have high bulk

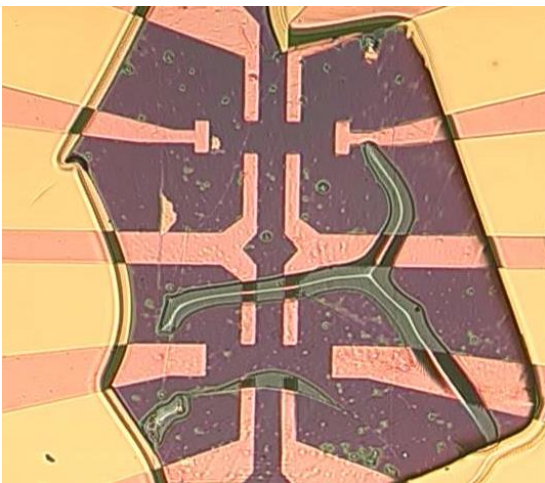


Fig.2 Optical micrograph of the sample. Thin single crystal was placed between the PDMS/Neoprim substrate and 4-terminal LCE. Each LCE have 10 electrodes in the single thin sheet of parylene film. 10 electrodes include drain, source, and electrical potential difference measurement electrodes. Gate electrode is not formed in this sample.

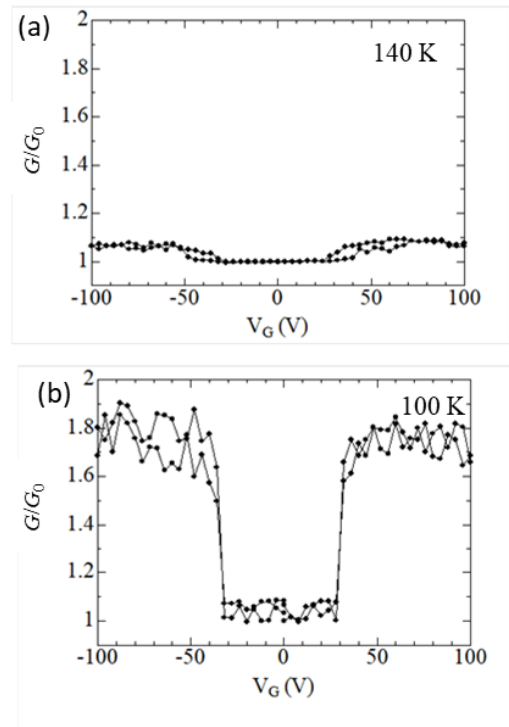


Fig. 3 Summary of the Temperature dependence of gate electric field-induced metal-insulator transition shown as relative electrical conductance (G/G_0) to applied gate voltage (V_G).

electrical conductivity of $10^2 - 10^{-1}$ S/cm even in the insulator phase. Moreover, Fig. 3 (a) showed obvious hysteresis behavior with applied gate voltage. The same V_G dependence is observed in Fig. 3 (b) at 100 K although the gate induced increase of G/G_0 become larger and the hysteresis behavior with V_G become smaller with decreasing temperature. Especially in Fig. 3 (b), saturated G/G_0 reaches to approximately 1.8 and V_G threshold converged to 28 V in the positive V_G side and -32 V in the negative V_G side, respectively. Abrupt increase and saturation of the relative conductance above the threshold voltage is distinctive features of the field induced phase transition FETs driven by dissolving highly correlated electronic insulator phase[2].

3. Conclusions

We have successfully discovered gate induced metal-insulator transition of charge order phase. Effectiveness of LCE even in low temperature is also clearly demonstrated.

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