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Article

Gate-Induced Thermally Stimulated Current on the Ferroelectric-like Dielectric Properties of (BEDT-TTF)(TCNQ) Crystalline Field Effect Transistor

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Abstract: A gate-induced thermally stimulated current (TSC) on β'-(BEDT-TTF)(TCNQ) crystalline FET were conducted to elucidate the previously observed ferroelectric-like behaviors. TSC which is symmetric for the polarization of an applied V_G^P and has a peak at around 285 K was assigned as a pyroelectric current. By integrating the pyroelectric current, temperature dependence of the remnant polarization charge was obtained and the existence of the ferroelectric phase transition at 285 K was clearly demonstrated. We have tentatively concluded that the phase transition between dimer Mott insulator and charge ordered phase occurred at around the interface of organic crystal and substrate.

Keywords: ferroelectricity; pyroelectric current; field effect transistor; Mott insulator; charge order

1. Introduction

Recently electronic ferroelectricity derived from charge ordered phase attract much attention in the wide range of the research area. In the quarter-filled charge ordered phase, carriers are localized

by on-site and inter-site Coulomb repulsion and form a stripe pattern of carrier distribution. The carrier patterns make large coherent domains in low temperature [1–6]. Since this is a kind of an electrically polarized electronic state, the charge distribution is modulated by an external electric field with increasing temperature. In these days, it is recognized that several types of organic correlated electronic materials exhibit electronic ferroelectricity.

We have investigated the field effect transistor (FET) characteristics of β'- (BEDT-TTF)(TCNQ) being known as an organic dimer Mott insulator [7,8]. The FET exhibited ambipolar FET characteristics at room temperature [9,10]. However, field effect mobility of both electron and hole abruptly changed at around 285 K and also showed the hysteresis on the temperature sweep [10]. We also found that the transfer curve of the FET indicated hysteresis on the applied gate voltage sweep below 285 K [10]. By the measurement of the temperature dependence and applied gate voltage dependence of gate capacitance included in a FET structure, we have found out that a ferroelectric-like transition exists in β'- (BEDT-TTF)(TCNQ). However, there has been no report on a ferroelectric-like phase transition observed in β'- (BEDT-TTF)(TCNQ) crystal. In addition, there has been no report on the ferroelectric-like phase transition at 285 K for two known polymorph [11–13]. To elucidate the origin of the ferroelectric-like properties, a thermally stimulated current (TSC) method was conducted on the FET sample.

TSC is a conventional electrical measurement to investigate a carrier trap and thermal dissolution of a spontaneous polarization. In this work, we conducted a gate-induced TSC measurement to mainly investigate a pyroelectric current. By measuring a pyroelectric current, the existence of spontaneous polarization which is the origin of the observed ferroelectric-like behavior is experimentally demonstrated.

2. Experimental Details

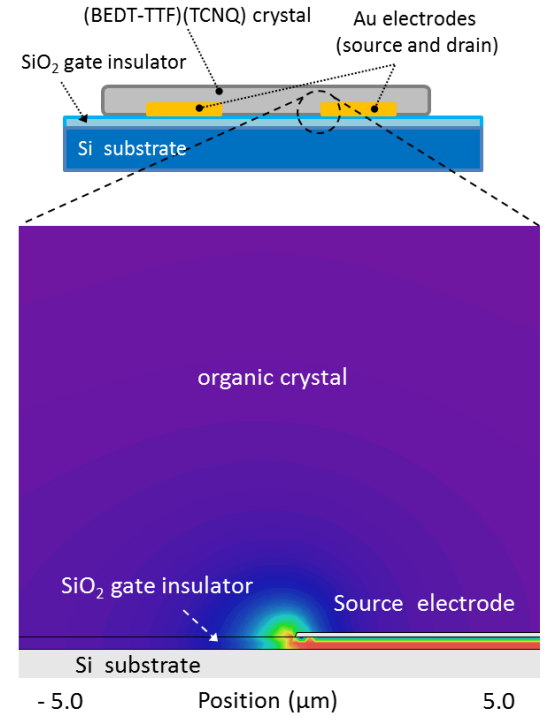
A Si wafer was cleaved and cleaned by warm semiconductor-grade acetone and UV/O_3 cleaner to remove organic contaminants on the surface. Then the Si wafer was cleaned by a semiconductor-grade H_2SO_4 mixture. Pre-fabricated surface SiO_2 layer of 1 μm thickness to protect the Si wafer was completely etched with a hydrofluoric acid mixture to remove alkali ion possibly diffused into SiO_2 layer. Then, SiO_2 layer of 300 μm thickness as a gate insulator of FET structure was formed by thermal oxidization. Then interdigital source and drain electrode of FET were fabricated by a standard vacuum evaporation method. The FET substrate was carefully prepared in a clean room.

The source materials, substrate preparation, and crystal growth method were described in our previous studies [9,10]. The high crystallinity, majority of the triclinic phase, and spontaneous oriented growth in the a^*-direction normal to the substrate surface were elucidated by the X-ray diffraction [9]. As a result, the $b-c$ conductive plane of the BEDT-TTF layer is parallel to the SiO_2 surface and continuous from source to drain electrode. A sample structure is not a normal capacitor structure but a bottom-contact FET structure itself because this ferroelectric-like phenomena is observed in this FET structure. In addition, dimension of the tiny crystal ($0.8 \times 300 \times 0.4$ μm) is not suitable for making parallel capacitor structure.

A gate-induced thermally stimulated current was measured in the procedure explained below. Initially, the sample was placed in vacuum chamber in which a base pressure is approximately 10^{-8} Pa and sample temperature was decreased to 280 K which is lower than the observed ferroelectric-like phase transition temperature (285 K). A poling voltage (V_G^P) was applied under 280 K and this poling bias was kept for

30 min. After the poling, the sample was cooled until about 90 K under the application of V_G^P to freeze the gate-induced polarization. After V_G^P was turned to 0 V, an electrical current from source electrode was measured under the constant sample heating rate of about 5 K/min. A thermally stimulated current was observed with the dissolution of initially frozen polarization.

Figure 1. Calculated electrical flux density distribution in the FET sample with the source and gate voltage of 0 and 30 V, respectively. The electric flux density in (BEDT-TTF)(TCNQ) is not uniform; far from that of parallel capacitor. The electric flux density is concentrated in the vicinity of the source electrode edge and at the interface between (BEDT-TTF)(TCNQ) and SiO_2 gate insulator.



3. Results and Discussion

At first, distribution of the electric field in our sample was calculated by the finite element method (FEM). Since the anisotropic dielectric constant of (BEDT-TTF)(TCNQ) is unknown, we assumed an isotropic dielectric constant of 100–10000 as trial values. This model dielectric isotropy is not so realistic because the 2-dimensional BEDT-TTF materials exhibit large anisotropy. However, the variation of dielectric constant barely affected the result of the FEM calculation. Figure 1 is a calculated distribution of an electric flux density in our bottom contact FET structure at around the source electrode. The thickness of source electrode is approximately 20 nm and the height of the organic crystals is ranging from 200 to 600 nm along a^* -axis. As shown in the figure, the electric flux density in (BEDT-TTF)(TCNQ) is not uniform but concentrated in the vicinity of the Au electrode edge and at the interface between (BEDT-TTF)(TCNQ) and SiO_2 gate insulator. The fringe field in the crystal is mainly consisted of a horizontal component which is parallel to the conductive b-c plane of BEDT-TTF layer.

Therefore, the main contribution to the observed dielectric response is due to the horizontal component of the dielectric constant of (BEDT-TTF)(TCNQ).

Dielectric constant estimated from the simulated capacitance was larger than the typical value measured in inorganic ferroelectric ceramics such as $BaTiO_3$. This estimation is tentative because of the limited precision of this calculation. However, early reports on one-dimensional charge ordered materials such as TMTSF compounds [14–19] indicated the extremely high dielectric constants of over 10000. Charge ordered state is one of the polarized state of which the polarization originate from the alternative localization of carrier. For the rigid charge ordered phase, it is impossible to invert the polarization by an external electric field. However, in a fluctuated charge ordered state, weakly localized carriers are mobile by the external electric field. Therefore, the effective electric field contributing to the observed dielectric response is horizontal component in the vicinity of the Au electrode and at the interface between (BEDT-TTF)(TCNQ) crystal and SiO_2 gate insulator, by no means from whole bulk.

Figure 2. Temperature dependence of the capacitance (C_p and dielectric loss (G) measured in (BEDT-TTF)(TCNQ) FET structure. C_p and G were measured by impedance meter at 4 Hz.

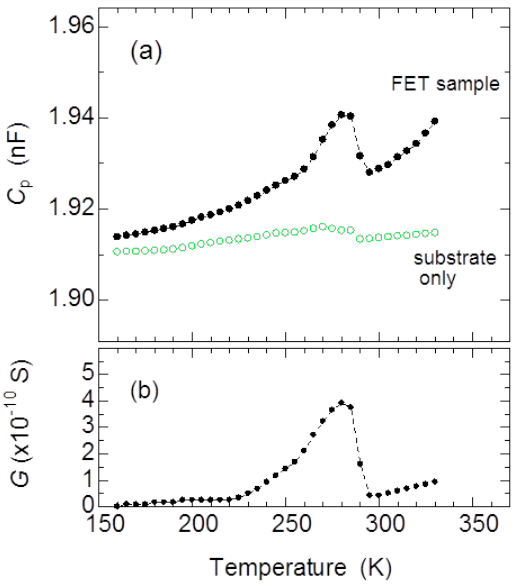


Figure 2 shows the temperature dependence of the dielectric response measured in (BEDT-TTF)(TCNQ) FET structure. C_p and G stand for a capacitance and a dielectric loss, respectively. The observed admittance is written as,

$$Y = G + j\omega C_\mathrm{p} = j\omega C_0 \left(\frac{C_\mathrm{p}}{C_0} - j\frac{G}{\omega C_0} \right) = j\omega C_0 \epsilon_\mathrm{r}^*$$

$$\epsilon_\mathrm{r}^* = \left(\frac{C_\mathrm{p}}{C_0} - j\frac{G}{\omega C_0} \right)$$

where j is the imaginary unit and C_0 is a geometrical capacitance. Therefore, C_p and G are related to the complex dielectric constant $\epsilon_\mathrm{r}^* = \epsilon_\mathrm{r}' + j\epsilon_\mathrm{r}''$. However in our case, it is impossible to transform C_p and G

into the ϵ'_r and ϵ''_r because an electric field is not uniform in the FET structure (see Figure 1). Although a parallel capacitor structure is generally used for conventional dielectric measurements, we had to observe a novel ferroelectric-like property which was observed in the channel region of FET structure [10] and had not been reported in bulk crystal. Both the observed C_p and G have a peak at around 280 K, and abruptly decrease at around 290 K. In our previous work [10], we suggested the existence of unknown ferroelectric-like transition by a displacement current measurement. In this work, the conventional impedance measurement also discovered this ferroelectric-like transition. However, compared to the typical ferroelectric transition, the observed temperature dependence of dielectric response is rather dispersive because the polarization in our sample is not uniform due to the non-uniformity of electric field in the crystal.

Figure 3. (**a**) Gate-induced TSCs observed in the FET sample after various V_G^p; (**b**) The TSCs observed in the substrate without (BEDT-TTF)(TCNQ) crystals.

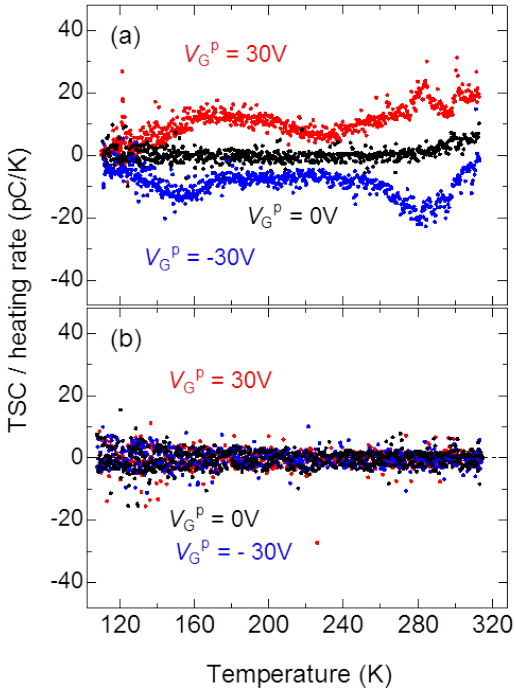
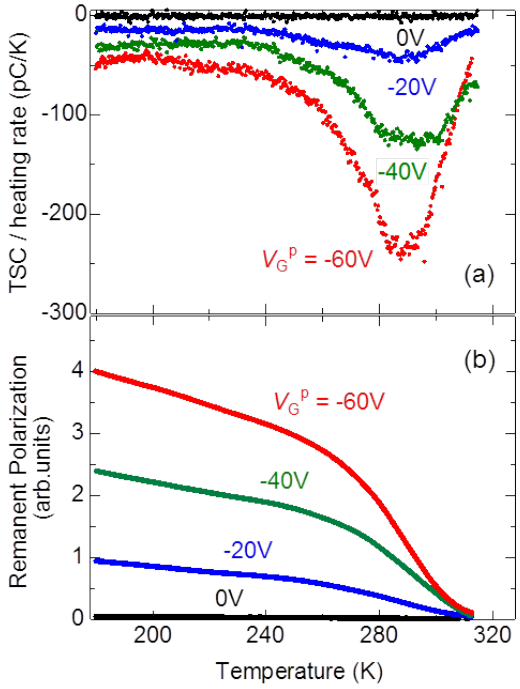


Figure 3(a) shows observed gate-induced TSCs. By application of the positive V_G^p, positive TSC was observed. On the other hand, negative TSC was observed by application of negative V_G^p. In addition, by application of zero V_G^p, no TSC peak was observed. Only a minor increase of a TSC was observed above 290 K, which was also observed in a blank test which is described afterwards. The observed TSC under non-zero V_G^p have a maximum at 285 K, and there are minor and broad peaks in 180–210 K. These minor and broad peaks do not belong to a pyroelectric current but correspond to detrapping of carriers because these peaks are not symmetric to the polarization of an applied V_G^p. A thermal activation energy for electron and hole detrapping should be different. On the other hand, major TSC observed above 210 K belongs to a pyroelectric current because these peaks are symmetric to the V_G^p polarization, and the TSC corresponds to the dielectric response and $Q-V$ hysteresis observed in our previous work [10].

The $Q - V$ hysteresis, divergent increase of dielectric constant and loss are a part of the feature of the ferroelectricity. Therefore, we concluded that the observed TSC corresponded to the dissolution of the ferroelectric-like polarization.

Here, other possible origin of observed TSC should be carefully considered. A possibility for mobile alkali ion (especially Na^+ ion in our environment) diffused into the SiO_2 gate insulator or a residual moisture on the substrate surface are denied by blank test in Figure 3 (b). Figure 3 (b) shows a result of blank tests which is measured by using the FET substrate without (BEDT-TTF)(TCNQ) crystals. There is no TSC signal in this sample. Although weak increase of TSC above 290 K was observed in some samples, the weak TSC did not depend on applied V_G^p and its polarity. In addition, the observed weak TSC decreases after the several thermal cycles. A possible origin of the weak TSC in the blank test is surface moisture or mobile alkali ion naturally diffused into the SiO_2 layer. However, thick pre-fabricated SiO_2 layer to protect the Si wafer from the alkali ion pollution is completely removed in the sample preparation procedure. Moreover, a contribution of alkali ion cannot decrease after several thermal cycles because alkali ion diffused in Si cannot vaporize in this experimental condition. Therefore, we conclude that the origin of the small TSC observed in the blank test is residual surface moisture at the substrate surface. Incidentally, the existence of ferroelectric phase of H_2O is proposed at around 60 K [20]. However, this phase cannot affect our results.

Figure 4. (**a**) V_G^p dependence of the observed TSCs; (**b**) Temperature and V_G^p dependence of the remnant polarization charge. The values for the remnant polarization charge at the lowest temperature (Q_{ini}) was taken rather arbitrarily to let the remnant polarization charge be proportional to V_G^p because it was difficult to determine the absolute value of Q_{ini} due to the non-uniform electric field.



Now we discuss the TSC as a pyroelectric current. Figure 4 (a) shows the $V_{\mathrm{G}}^{\mathrm{p}}$ dependence of the observed TSCs. These TSCs increase with increasing $V_{\mathrm{G}}^{\mathrm{p}}$ because induced average polarization increase with increasing $V_{\mathrm{G}}^{\mathrm{p}}$. However, the TSC peak temperature shows little dependence on the $V_{\mathrm{G}}^{\mathrm{p}}$. In this temperature region, the observed TSC is expected to mainly consist of the pyroelectric current because the temperature range is higher than that of which the detrapping from the carrier traps are observed. On this basis, we integrated the observed TSCs. The relation between the polarization and observed TSC is written as

$$i(T) = -\frac{dQ_{\mathrm{r}}}{dt} = -\frac{dQ_{\mathrm{r}}}{dT}\frac{dT}{dt} = -s_{\mathrm{eff}}\frac{dP_{\mathrm{r}}}{dT}\frac{dT}{dt}$$

where, s_{eff} is the effective surface area of the sample and is defined for a parallel capacitor. Q_{r} and P_{r} is a remnant polarization charge and a remnant polarization, respectively. In the case of the parallel capacitor, one can define the pyroelectric coefficient p(T),

$$\frac{dP_{\mathrm{r}}}{dT} = p(T)$$

thus one obtains:

$$i(T) = -s_{\mathrm{eff}}p(T)\frac{dT}{dt}$$

If the heating rate ($\frac{dT}{dt}$) is constant, one can obtain the pyroelectric coefficient. Then, P_{r} was calculated as an integration of the pyroelectric current $i(T)$.

$$P_{\mathrm{r}}(T) = -\int p(T)dT = -\frac{1}{s_{\mathrm{eff}}}\int \frac{i(T)}{\frac{dT}{dt}}dT$$

In our case, remnant polarization charge is calculated as,

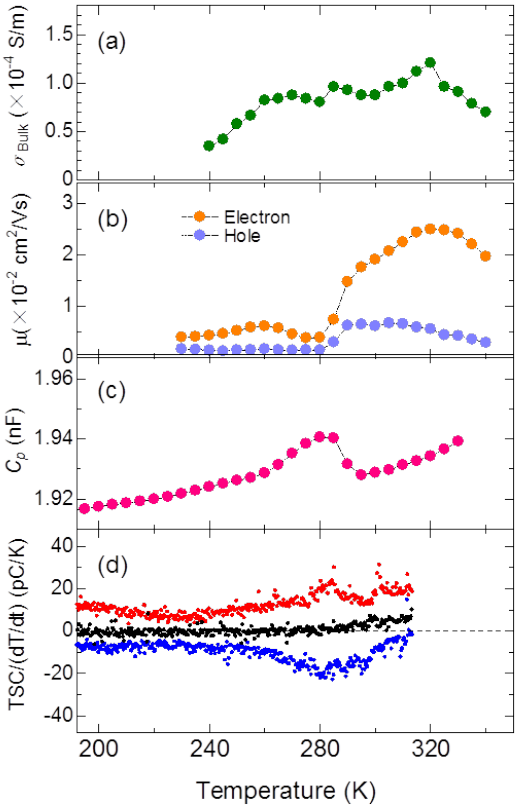
$$Q_{\mathrm{r}}(T) = -\int \frac{i(T)}{\frac{dT}{dt}}dT + Q_{\mathrm{ini}}$$

where Q_{ini} is a constant of integration which is fixed by an initial condition, *i.e.*, initially induced polarization. Figure 4 (b) shows integrated TSC, which corresponds to the temperature and $V_{\mathrm{G}}^{\mathrm{p}}$ dependence of the remnant polarization charge (Q_{r}). Although the Q_{ini} is difficult to determine, because of the non-uniform electric field in the crystal, we determined the Q_{ini} for each $V_{\mathrm{G}}^{\mathrm{p}}$ on the assumption that the initially induced Q_{r} is proportional to the $V_{\mathrm{G}}^{\mathrm{p}}$. Figure 4 (b) indicates that Q_{r} gradually decrease with increasing temperature until 260 K, and begin to decrease steeply at around 280–290 K. A little Q_{r} remains at 320 K. These observed temperature dependence well corresponds to the temperature dependence of the C_{p} as shown in Figure 2 (a). Temperature dependence of C_{p} and previously observed hysteresis [10] had suggested the existence of ferroelectric phase transition, which is now supported by the additional evidence in this work.

Figure 5 are the summary of the physical and electrical parameters of the β'-(BEDT-TTF)(TCNQ) crystalline FET. There are clear relationships between the observed parameters; especially, the ferroelectric-like transition, which is indicated in Figure 5 (c) and (d), and a change of the field effect mobility (shown in Figure 5 (b)) occurred simultaneously. On the other hand, this ferroelectric-like transition hardly affected on the bulk conduction shown in Figure 5 (a). Therefore, this correlation

is related to the interface of the organic crystals and SiO_2 because both field effect mobility and the ferroelectric-like properties were observed at around the interface, not as the bulk properties. Because a bulk β'-(BEDT-TTF)(TCNQ) crystal is known as a dimer Mott insulator, the ferroelectric-like phase below 285 K, discovered in this work, is exceptional. On the other hand, there are many examples of ferroelectric phase transition observed in charge ordered materials [1–6]. Therefore, we have speculated that the phase transition between the charge-ordered phase and dimer Mott insulator phase occurred at the interface of organic crystal and gate insulator.

Figure 5. Summary of the physical and electrical parameters observed in the β'-(BEDT-TTF)(TCNQ) crystalline FET. Temperature dependence of (**a**) bulk conductance; (**b**) field effect mobility; (**c**) gate capacitance, and (**d**) TSC.



The band filling of both the dimer Mott insulator and charge ordered phase are quarter filling. The difference of the dimer Mott insulator and charge ordered phase is the degree of localization of correlated carriers. The carrier of the dimer Mott insulator and charge ordered phase is localized in one dimer site and one BEDT-TTF site, respectively. If a carrier is localized in a BEDT-TTF site and the thermal hopping to the intradimer neighboring BEDT-TTF site is allowed, spontaneous polarization derived from the biased carrier distribution in dimer sites is induced by an external electric field. Based on this assumption, the strange temperature behavior shown in Figure 5 is explained by the abrupt change of the degree of the localization on correlated carriers. And also, phase transition between charge ordered phase and Mott insulator phase in a quarter-filled dimer Mott insulator had been predicted in theoretical

works [21–23]. By introducing the intradimer strain, Mott insulator phase undergoes the phase transition to ferroelectric phase in the way to the charge ordered phase.

Although the effect of a gate electric field on the phase transition is not clear now, it is indubitable that an interface strain on the soft organic material affects this phase transition. A modulation of a crystal lattice parameter is one of the necessary factor to lead the phase transition between charge ordered and Mott insulator phase in the theoretical prediction [21–23]. Thermal expansion coefficient of Si wafer is much smaller than that of the organic materials. Therefore, if there is no slippage at the interface, the in-plane (b-c plane) lattice parameter of the organic crystal at around the interface of the Si wafer is effectively expanded with decreasing temperature compared to its thermal expansion in free crystal [24,25]. In fact, in our preliminary result, distance of $(0kl)$ planes are expanded compared to the free $(0kl)$ plane distance (to be published elsewhere). On the other hand, $(h00)$ plane distance is rather compressed compared to $(h00)$ distance of the free bulk crystal. These facts are explained by the interfacial strain on the soft organic material.

4. Conclusions

We have presented the gate-induced TSC experiment on (BEDT-TTF)(TCNQ) crystalline FET sample to elucidate previously observed ferroelectric-like behaviors and related abrupt change of the field effect carrier mobility. Among the observed TSC components, TSC which is symmetric for the applied $V_{\mathrm{G}}^{\mathrm{p}}$ and has a peak at around 285 K was assigned as a pyroelectric current derived from the spontaneous polarization of ferroelectricity. By integrating the pyroelectric current, temperature dependence of the remnant polarization charge was exhibited and the existence of the ferroelectric phase transition at 285 K was clearly demonstrated. With the calculated distribution of an electric field and obvious relationship of the field effect mobility and ferroelectric behavior, we have concluded that the observed novel phenomena is brought about at around the interface of organic crystal and substrate. Although the phenomena has not fully been elucidated yet, we temporarily concluded that the phase transition occurred between dimer Mott insulator and charge ordered phase.

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