

Studies of Field-Effect-Transistor Based on Highly-Correlated Molecular Materials

Department of Materials Molecular Science
Division of Electronic Properties



YAMAMOTO, Hiroshi
URUICHI, Mikio
SHIMIZU, Atsuko

Professor (April, 2012–)
Technical Associate
Secretary

Field effect transistor (FET) with organic molecules as a channel material is under intense studies because of its application possibilities such as flexible, printable, and large-area electronic devices. Despite such a thorough investigation on single-component neutral molecules to pursuit high performance (say, high mobility *etc.*) in FET uses, few studies are known for compound organic semiconductor-based transistors. We have been exploiting FET devices with charged molecular materials, namely cation-radical salts of electron-donating molecules (donors) such as BEDT-TTF and anion-radical salts of electron accepting molecules (acceptors) such as Ni(dmit)₂. Among these materials, our focus concentrates in Mott-insulators in which Coulomb interaction among carriers blocks metallic transport. In this highly correlated situation of charge carriers, Mott insulator stays in a fragile semi-conducting state, where carrier injection, chemical and/or physical pressure (or strain), and thermal fluctuation can drive it into a metallic state by phase transition (*i.e.* Mott transition: Figure 1). Since the carrier density (band-filling) of a FET interface can be finely modulated by an electrostatic field from the gate electrode, it is anticipated that the Mott-insulating state can be switched to a metallic state by a field effect.

Indeed, our previous work showed such an insulator-to-metal transition in terms of the band structure, and these results provide not only a possibility of application use of organic Mott-FET but also a significant insight into the mechanism of Mott transition itself. We are now expanding our research target both to the superconducting transition of Mott-FET and to its room-temperature operation.

(BEDT-TTF = bis(ethylenedithio)tetrathiafulvalene, dmit = 1,3-dithiole-2-thione-4,5-dithiolate)

1. Field-Induced Superconductivity in an Organic Mott-FET

κ -(BEDT-TTF)Cu[N(CN)₂]Br (κ -Br) is an organic superconductor whose electronic state is Mott-insulating at room-temperature but turns into metallic at low temperature through a crossover around 50–100 K, possibly because of an increase of bandwidth upon thermal contraction. In our previous works, a tensile strain altered its ground state into a Mott-insulating state, when its thin (100–300 nm) crystal is laminated on top of SiO₂/Si⁺⁺ substrate and cooled down to low temperature.

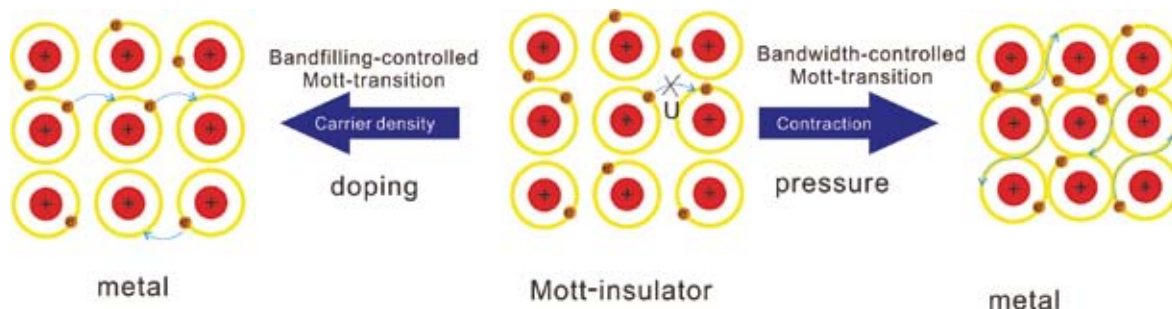


Figure 1. Concept of a Mott-insulator (center) along with its bandfilling-controlled (left) and bandwidth-controlled (right) Mott transitions. When the band is half-filled and band energy is small, the carriers are localized because of on-site Coulomb interaction (U). This situation is broken down either by changing the carrier concentration or by compressing the lattice. In many cases, the Mott transitions are associated with superconductivity at the boundary between metallic and insulating phases.

Although the electronic state at low temperature became completely insulating in this experiment because of the very small thermal expansion coefficient of Si substrate (2 ppm/K), one can anticipate from the T - P (temperature vs. pressure) phase diagram that somehow mixed electronic state between superconducting and Mott-insulating states can be realized when the tensile strain is much weaker. To achieve such a mixed state in the device, where phase-separation occurs between superconducting and Mott-insulating states, we have chosen Nb-doped SrTiO₃ as a back-gate substrate because of its larger thermal expansion coefficient (*ca.* 10 ppm/K) than Si. An aluminum oxide layer was grown by atomic layer deposition technique to form a gate dielectric on the substrate. After lamination process of κ -Br on the substrate, the Mott-FET device which showed a weakly insulating behavior at low temperature was fabricated (Figure 2). Upon applying a positive gate voltage, however, the resistivity goes down and weakly metallic behavior was observed at $V_G > 2$ V. By further increasing the gate voltage up to 8 V, the device showed a sudden drop of resistivity around 5 K, which can be attributed to superconductivity. Taking account of bistable IV characteristics observed in the low resistance region, the above transition can be understood as a percolation transition of superconducting islands that is induced by the electrostatic doping of electrons. The transition temperature increases as the gate voltage rises and saturates around $V_G = 11$ V. The above result is the first example of field-induced superconductivity in organic materials, and can be utilized for uncovering a phase diagram of organic Mott system in the band filling controlled regime.

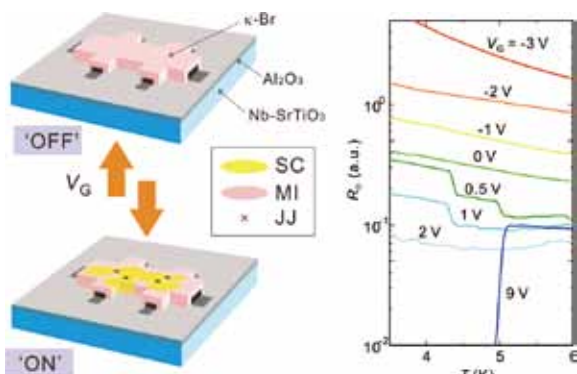


Figure 2. The device structure and temperature dependency of device resistance under various gate voltages. The device comprises κ -Br thin-layer single crystal and Al₂O₃/SrTiO₃ substrate. By applying a positive gate voltage, the resistance of the device starts decreasing, while a negative gate voltage results in an increase of the resistance. Although superconducting (SC) and Mott-insulating (MI) phases are separated in the device, the four-terminal resistance drops to zero when the Josephson junction (JJ) network forms a pathway to shunt the voltage terminals. This kind of percolation transition is clearly observed in the right panel, where the resistance drops at 5 K with a gate voltage of 9 V.

2. Operation of Organic Mott-FET at Higher Temperature

In order to operate the organic Mott-FET at higher temperature, hopefully at room temperature, it is necessary to realize much larger Mott gap energy and thinner crystal thickness. For this purpose, we are examining several strategies in parallel.

κ -(BEDT-TTF)Cu[N(CN)₂]Cl (κ -Cl) is a Mott-insulator with larger Mott gap than κ -Br and is therefore suitable for examining the influence of an increase in Mott-gap energy on the device performance. For example, it has higher resistance than κ -Br and exhibit better ON/OFF ratio. In addition, it always shows ambipolar behavior, probably because of its clean surface (Figure 3). This situation allows us to analyze the critical exponents of filling-controlled Mott transition, which will give us important information about ON/OFF efficiency of the device. Because it is also possible to control the Mott gap energy by applying a mechanical strain, κ -Cl device provides an ideal platform to expand Mott-FET strategy to higher temperature.

Another candidate of the channel material for Mott-FET that can be used at room temperature is (BEDT-TTF)(TCNQ). We are exploiting this material both in a crystalline and amorphous form to find a better FET setup for simple but highly efficient device at room temperature. (TCNQ = tetracyanoquinonodimethane)

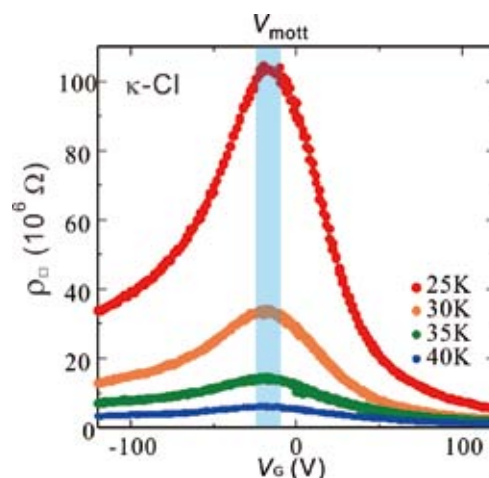


Figure 3. Gate voltage dependence of the resistance for κ -Cl based Mott-FET. Because this device clearly shows ambipolar behavior due to a smaller number of mid-gap trap states, it is possible to exactly define the gate voltage for charge neutrality point (V_{Mott}). This fact helps determining the critical exponent for Mott transition, which is important both for theoretical investigation and for analyzing device parameters such as subthreshold swing.

Reference

- 1) Y. Kawasugi, H. M. Yamamoto, N. Tajima, T. Fukunaga, K. Tsukagoshi and R. Kato, *Phys. Rev. B* **84**, 125129 (9 pages) (2011).