Open up Future Electronics by Organic Molecules

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Education

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Professional Employment

- 1998 Research Associate, Gakushuin University
- 1999 Special Postdoctral Fellow, RIKEN
- 2000 Research Scientist, RIKEN
- 2007 Senior Research Scientist, RIKEN
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Awards

- 2009 RSC Publishing CrystEngComm Prize
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Organic molecules are attracting recent attention as new ingredients of electronic circuits. Their functionalities have been developed considerably, but are still to be explored and advanced. Our group focuses on a development of organic electronics in the next era by providing new mechanism and concepts of the device operation and fabrication. For example, an electronic phase transition is utilized for the ON/OFF switching of our field-effect-transistor (FET). This special FET is called an organic Mott-FET, where the conduction electrons in the organic semiconductor are solidified at the OFF state because of Coulomb repulsion among carriers. However, these solidified electrons can be melted by applying a gate voltage, and show an insulator-to-metal transition so-called Mott-transition to be switched to the ON state. Because of this phase transition, a large response of the device can be achieved, resulting in the highest device mobility ever observed for organic FETs. At the same time, Mott-transition is known for its relevance to superconductivity. Not only in organic materials but also in inorganic materials such as cuprates, Mott-transition is frequently associated with superconducting phase at low temperature. Indeed, our organic FET shows an electric-field-induced superconducting transition at

Selected Publications

- H. M. Yamamoto, M. Nakano, M. Suda, Y. Iwasa, M. Kawasaki and R. Kato, "A Strained Organic Field-Effect Transistor with a Gate-Tunable Superconducting Channel," *Nat. Commun.* 4, 2379 (7 pages) (2013).
- H. M. Yamamoto, "Sheathed Nanowires Aligned by Crystallographic Periodicity: A Possibility of Cross-Bar Wiring in Three-Dimensional Space," *CrystEngComm* 16, 2857–2868 (2014).
- Y. Kawasugi, H. M. Yamamoto, N. Tajima, T. Fukunaga, K.

low temperature.

Another approach to the future electronics is a threedimensional (3D) patterning of molecular devices using crystal engineering. Because each molecule can be designed to show different functionalities, it should be attractive to construct nano-structured devices by self-assembly. We are especially focusing on a development of supramolecular nanowires that allow 3D periodic wiring in nano-scale. By encapsulating a 1D array of conducting molecules in a channel formed inside 3D supramolecular network, it is possible to construct a sheathed nanowires aligned in a periodic order as shown in Figure 1.



Figure 1. Crystal structure of supramolecular nanowire.

Tsukagoshi and R. Kato, "Field-Induced Carrier Delocalization in the Strain-Induced Mott Insulating State of an Organic Superconductor," *Phys. Rev. Lett.* **103**, 116801 (4 pages) (2009).

 H. M. Yamamoto, Y. Kosaka, R. Maeda, J. Yamaura, A. Nakao, T. Nakamura and R. Kato, "Supramolecular Insulating Networks Sheathing Conducting Nanowires Based on Organic Radical Cations," *ACS Nano* 2, 143–155 (2008).

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1. Field-Induced Superconductivity in an Organic Mott-FET¹⁾

κ-(BEDT-TTF)Cu[N(CN)2]Br (κ-Br) is an organic Mottinsulator at room-temperature, but turns into metallic and superconducting states at low temperature. In our previous works, a tensile strain from FET substrate altered its ground state into a Mott-insulating state, when its thin (100-300 nm) crystal was laminated on top of SiO2/Si++ substrate and cooled down to low temperature. In those experiments the electronic state at low temperature became completely insulating because of the tensile strain that originates in mismatching of thermal expansion coefficients between K-Br (30 ppm/K) and Si substrate (2 ppm/K). However, one can anticipate from the T-P(temperature vs. pressure) phase diagram that 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 (or, percolate-superconducting 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 of κ -Br on the substrate, the Mott-FET device which showed a weakly insulating behavior at low temperature was fabricated (Figure 2).

Upon application of a positive gate voltage, the resistivity goes down and weakly metallic behavior is observed at $V_{\rm G} > 2$ V. By further increasing the gate voltage up to 8 V, the device shows a sudden drop of resistivity around 5 K, which can be attributed to superconductivity. Taking into 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 $V_{\rm G} = 11$ V. This 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 simultaneous control of band filling and band width.

2. Strain-Tunable Organic FET with Strongly Correlated Electron Systems²⁾

Organic materials are soft and can be stretched even at low temperature. Our organic Mott-FETs are also soft enough to be compressed or expanded with a plastic substrate and show a band-width-controlled Mott/superconducting transitions. This kind of lattice modulation has been demonstrated in our recent experiments by mechanically pushing a poly-ethylene naphthalate (PEN) substrate covered with gold gate electrode and parylene-C dielectric from the back side (Figure 2). The organic Mott-insulator κ-(BEDT-TTF)Cu[N(CN)₂]Cl (κ-Cl) laminated on top of the substrate exhibited superconductor-toinsulator transition by bending the substrate where the surface has an effect of tensile strain in a longitudinal direction. The strain necessary for this transition is only 0.3% while the resistance change reached ten orders of magnitude, which demonstrates high sensitivity of the Mott-insulator. The phase diagram obtained by simultaneous scans of strain and temperature well reproduces that of hydrostatic pressure. This means that one-dimensional tensile strain corresponds to an effective negative pressure in κ-Cl system.

Then, a field-effect was examined by applying a gate electric field. The transfer characteristics at higher tensile strain showed only moderate device mobility of around 5 cm²/Vs. When the system was forced to stay in a mixed phase state, where superconducting and Mott-insulating phases coexist, however, the electric-field-effect maximized to show device mobility up to 5900 cm²/Vs. This high response of the device can be attributed to an increase of superconducting fraction at application of gate voltage. In order to examine the above hypothesis, we have applied a magnetic field up to 7 T. Indeed, the field-effect has been considerably suppressed because the superconducting phases are destroyed by the magnetic field. This electric-field-induced superconductivity seems to originate from a decrease of Coulomb repulsion among carriers by electrostatic doping effect.

(BEDT-TTF = bis(ethyelenedithio)tetrathiafulvalene)



Figure 2. Device schematic (left) and temperature dependency of resistance at various gate voltages (right).



Figure 3. Side-view schematic for flexible organic Mott-FET device.

References

- 1) H. M. Yamamoto, M. Suda, M. Nakano, Y. Iwasa, M. Kawasaki and R. Kato, *Nat. Commun.* **4**, 2379 (7 pages) (2013).
- M. Suda, Y. Kawasugi, T. Minari, K. Tsukagoshi, R. Kato and H. M. Yamamoto, *Adv. Mater.* 26, 3490–3495 (2014).