Charge Dynamics at the Interface of Organic/Inorganic Composites: Best-performing Organic Field-effect Transistors Featuring Organic Crystalline Surfaces

Background

"God created the solids, the devil their surfaces" (W. Pauli). Since addressed in early 19's, a number of surface/interface properties still remain mysteries. On the other hand, we are being profited by technologies such as batteries and catalysts which utilize the charge motion at/around the interfaces. Therefore, CRIEPI-MSRL believes that microscopic understanding of the interface directly leads to development of next-generation energy technologies. Among varieties of material combinations, organic/inorganic interfaces are particularly promising, noticing sensitivity and controllability of the former and structural stability of the latter. We extract attractive functions of the unique composite by employing advanced techniques of charge-transport measurements established in our laboratory.

Objectives

To develop a method for fabricating ideal organic/inorganic interface making use of self-organization of the organic molecules, and to construct high-performance field-effect transistors with the self-organized high-quality interface.

Principal Results

1. Development of a method to fabricate an ideal organic/inorganic interface

In the newly developed method shown in Fig.2a, organic crystals are independently grown (Fig.1) and are softly attached by natural electrostatic force on SiO_2/doped Si substrates, whose surfaces are coated with organosilane self-assembled monolayers[*1]. Minimizing damages on the crystal surface, the soft lamination technique provides flat and homogenious organic/inorganic interfaces, which were inaccessible by conventional methods such as vacuum evaporation.

2. Pioneering measurements of charge transport at the surface of organic crystals with field-effect-transistor structures

In the field-effect transistors (FETs), charges are induced at the interface as shown in Fig.2b when the gate voltage is applied to the SiO_2 dielectric (just as the mechanism of capacitors). To examine quality of the single-crystal interface, we have developed a "four-terminal" measurement of the charge conductivity induced at the crystalline surface. The pioneering measurement revealed that amount of interface defects in the single-crystal FETs are much less than that in normal polycrystalline-based FETs. Pronounced mobility 9 cm^2/Vs of the interface charge conduction is realized as a result.

3. The best switching performance as a consequence of minimized interface defects

The best FET performance is achieved with a particular organic compound called Rubrene; on-off switching property due to opening of the positive charge-conduction channel (negative charge cannot transport in this system) is improved by 3 times as compared to best organic devices ever reported, owing to the minimized defect density as small as 10^{11} (a patent submitted). The result indicates that the self-organization of the organic interface is a promising strategy to have high-performance FETs, as well as to develop low-loss devices for energy conversion in the future.

Future Developments

In addition to the present study of the charge dynamics in parallel to the interface, we will extend the measuremnt to the charge transport across the interface. Microscopic elucidation of electronic and ionic conduction through metal-organic interfaces gives an indication for developing a new energy storage and/or conversion devices.

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Reference

"Field-induced charge transport at the surface of pentacene single crystals: a method to study charge dynamics of two-dimensional electron systems in organic crystals", J. Takeya et al., Journal of Applied Physics 94, 5800 (2003);
"Effects of polarized organosilane self-assembled monolayers on organic single crystal field effect transistors", J. Takeya et al., Applied Physics Letters 85, 5078 (2004).

＊1：organosilane self-assembled monolayer: lengthy molecules such as $CH_3(CH_2)_7(CH_2)_2Si(OC_2H_5)_3$, whose alkane chains are terminated with a silane group. The silane group chemically bonds the SiO_2 surface to form self-ordered film with the thickness of one-molecular length. The technique is actually applied to treat a glass surface to hydrophobic, for example.

＊2：Rubrene: An organic molecule ($C_{42}H_{28}$) used for yellow electroluminescence.

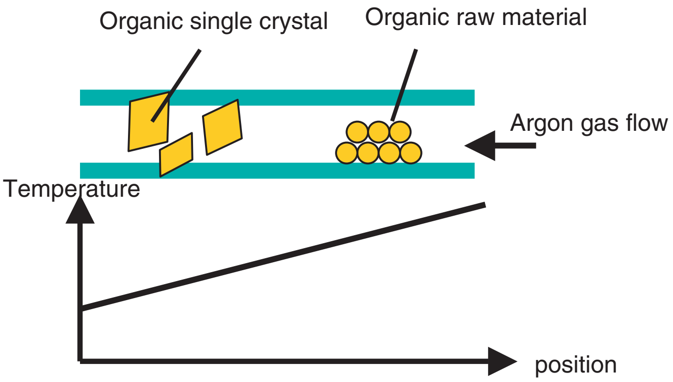

Fig.1 Physical Vapor Transport: a growth technique of organic single crystals

In the presence of temperature gradient of 200-300℃, organic molecules are sublimed at the higher-temperature right-hand side, carried by Ar gas, and crystallize at the lower-temperature left-hand side of the ftube furnace.

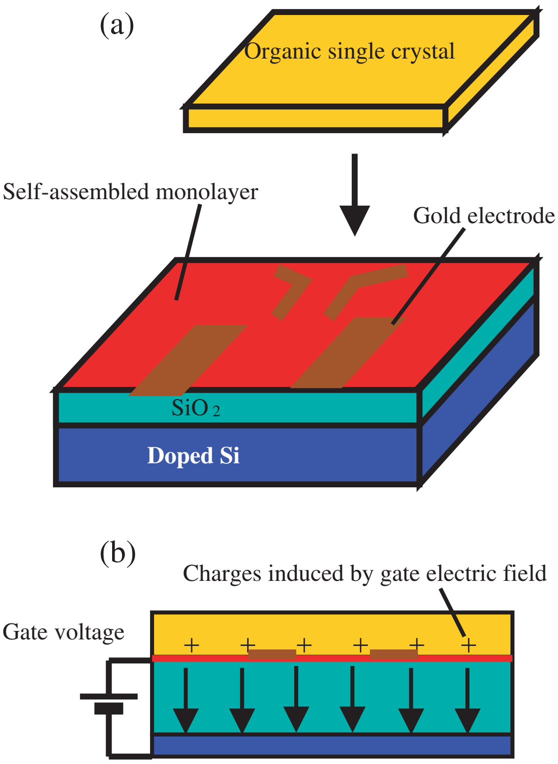

Fig.2 (a) Newly developed method to fabricate single-crystal field-effect transistors. (b) Mechanism of the charge accumulation by the gate-volatge application. Front view of the device depected in (a).

Charge is accumulated at the surface of the organic crystal in proportion to gate voltage VG, so that the interface current between the gold electrodes is amplified with V_G (field-effect). The extent of the current enhancement is determined by molecular-scale flatness of the interface.

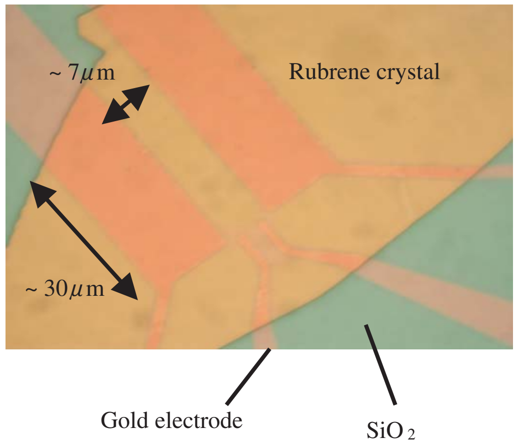

Fig.3 Top-view photo of the rubrene-single crystal

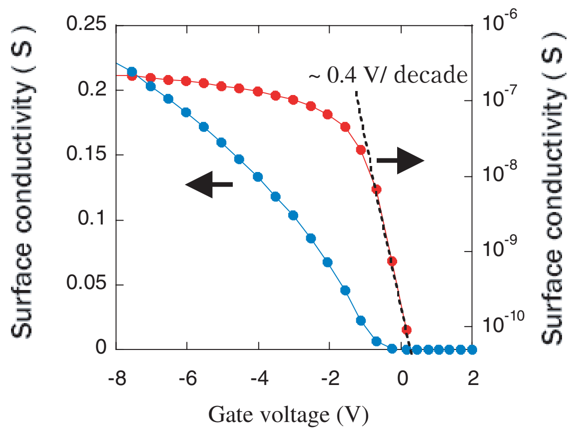

Fig.4 Outstanding switching performance of rubrene crystal/self-assembled monolayer/SiO_2 structure. Blue dots are plotted in a linear scale, and red are in alogarithmic scale.

With application of gate voltage, the transistor switches from off- to on-state with the slope of ~ 0.4 V/decade, demonstrating the highest-class organic FET performance. The steepness indicates successful fabrication of a near ideal interface of the self-organized interface. The steepness of the switch is essential in low-power applications such as for logic circuit components.