

Open up Future Electronics by Organic Molecules

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Organic molecules are attracting recent attention as new ingredients of electronic circuits. Our group focuses on the 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. In the operation, 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 electric response of the device can be achieved, resulting in the highest device mobility ever observed for organic FETs. In addition to this high performance, the Mott-FET is interesting in terms of superconductivity. Because the Mott-transition sometimes accompanies superconducting phase in between metal and insulator, modulation of gate electric field at low temperature may induce superconductivity. In fact, we have achieved first example of field-induced superconductivity in an organic FET. By combining a strain effect that can tune the bandwidth, this type of electric-field-induced superconducting transition can

be utilized for mapping the phase diagram around the Mott-insulator as shown in Figure 1.

Another approach to the future electronics is the development of spintronic devices based on chirality of organic material. We aim to implement chirality-induced spin selectivity (CISS) effect into molecular devices that can generate spin-polarized current. This type of device is expected to realize spintronics devices without magnet or topological insulator.

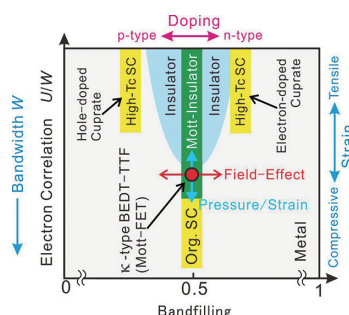


Figure 1. Phase diagram surrounding a Mott-insulator. SC denotes superconductor, while U and W are on-site Coulomb repulsion and bandwidth, respectively.

Selected Publications

- H. M. Yamamoto, "Phase-Transition Transistor Based on Organic Mott Insulators," *Bull. Chem. Soc. Jpn.* **94**, 2505–2539 (2021).
- Y. Kawasugi, K. Seki, S. Tajima, J. Pu, T. Takenobu, S. Yunoki, H. M. Yamamoto and R. Kato, "Two-Dimensional Ground-State Mapping of a Mott-Hubbard System in a Flexible Field-Effect Device," *Sci. Adv.* **5**, eaav7282 (9 pages) (2019).
- M. Suda, Y. Thathong, V. Promarak, H. Kojima, M. Nakamura, T.

Shiraogawa, M. Ehara and H. M. Yamamoto, "Light-Driven Molecular Switch for Reconfigurable Spin Filters," *Nat. Commun.* **10**, 2455 (7 pages) (2019).

- M. Suda, R. Kato and H. M. Yamamoto, "Light-Induced Superconductivity Using a Photo-Active Electric Double Layer," *Science* **347**, 743–746 (2015).

1. Current-Induced Spin-Polarization in a Chiral Crystal CrNb_3S_6 ^{1,2)}

CISS effect has remarkable ability which generates highly polarized spin current even with light element molecules. However, its extension to inorganic chiral materials has not been well investigated. Moreover, detection of CISS effect in metals that show ohmic response is quite interesting because one can discuss the CISS-based spin polarization in terms of band theory if metallic CISS effect in linear response regime is observed. So far, however, CISS experiments have been investigated only in tunnelling conduction regime.

We detected CISS-based spin transport phenomena in a monoaxial chiral dichalcogenide CrNb_3S_6 . This material has chiral structure and metallic conduction, so that we could perform CISS experiments with metallic conduction regime. Spin polarization was detected in this chiral bulk crystal under a charge current flowing along the principal c axis at room temperature without magnetic field. The detection was made by an inverse spin Hall signal which is induced on the tungsten electrode that absorbs polarized spin from the chiral crystal. An inverse response was also observed when applying the charge current into the tungsten electrode, which implied an inverse CISS effect. The signal sign reversed in the device with the opposite chirality, which is consistent with the symmetry required for CISS effect. Furthermore, the spin signals were found over micrometer length scale in a nonlocal configuration. Such a robust generation and protection of the spin-polarized state can be discussed based on a one-dimensional model with an antisymmetric spin-orbit coupling.

In addition to the above experiments, we also detected bulk magnetization generated by applying electric current to the crystal using SQUID magnetometer (Figure 2). When the current amplitude was swept from negative to positive, the current-induced magnetization changed linearly. Directly detecting such magnetization by magnetometry enables one to estimate the number of spin-polarized electrons. Using this

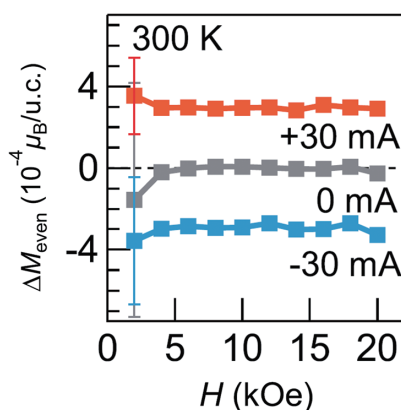


Figure 2. Detection of spin polarization in a chiral metal CrNb_3S_6 . By applying electrical current, electron spins are polarized along the current direction by CISS effect. The amplitude of the magnetization is irrelevant to the applied magnetic field, which strongly supports the current-induced nature of this magnetization.

number, we evaluated the spin polarization rate within the framework of Boltzmann's equation and found that spin polarization generated by CISS effect was enhanced by 10^5 times inside this material. It seemed that effective magnetic field generated by CISS could reach 10^3 T at high current density, which again confirmed the robustness of CISS effect. We also observed that the current-induced magnetization increased in the vicinity of the phase boundary between paramagnetic and forced ferromagnetic phases, which could be attributed to the spin fluctuation associated with the phase transition. (SQUID = superconducting quantum interference device)

2. Spin Current Generation in a Chiral Organic Superconductor

Although centrosymmetric s - and d -wave superconductors are in a spin singlet state, a superconductor with broken mirror symmetry is expected to show spin triplet state, according to a theory developed by Edelstein.³⁾ This means spin polarization can be generated by applying supercurrent in a chiral superconductor, whose magnetization direction depending on the lattice symmetry has been recently calculated by group theory.⁴⁾ We have tested this idea by employing κ -(BEDT-TTF)₂Cu(NCS)₂ (hereafter, κ -NCS) which is an organic superconductor with chiral and polar crystal lattice. The space group of this crystal is $P2_1$, and its handedness is defined by the relative arrangement between the anionic $\text{Cu}(\text{NCS})_2$ and cationic BEDT-TTF. This handedness can be experimentally determined by X-ray diffraction or circular dichroism. After confirming pure enantiomeric lattice system, a thin crystal of κ -NCS has been laminated onto a resin substrate with pre-patterned gold and nickel electrodes. At temperature lower than superconducting T_c , an electrical current was applied to induce spin magnetization. The spin polarization accumulated at the interface between κ -NCS and the magnetic electrode was detected as a voltage that is dependent on the magnetic field. By measuring the angle dependency of this magneto-voltaic signal, the direction of accumulated spin could be determined. The spin polarization direction was dependent on the specific location inside the crystal, and its arrangement was consistent with a magnetic quadrupole structure which has been hypothesized in a chiral molecule with CISS effect. [BEDT-TTF = bis(ethylenedithio)tetrathiafulvalene]

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