Charge coherence and electronic correlation in highly doped organic single-crystal double quantum wells

Jun Takeya,[1,2,3] Naotaka Kasuya,[1] Shun Watanabe,[1] Hideaki Takayanagi,[1] Daisuke Yamanaka,[1] Tomoki Furukawa,[1] Yu Yamashita,[1,3] Mizuki Abe,[1] and Toshihiro Okamoto[1,2]

[1] Graduate School of Frontier Sciences, The University of Tokyo, Kashiwa 277-8561, Japan, e-mail: takeya@k.u-tokyo.ac.jp
[2] CREST, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan
[3] MANA, National Institute for Materials Science, Tsukuba 305-0047, Japan

We introduce a novel two-dimensional hole system in small-molecular assembly with electronic phases continuously controlled from a wide-gap insulator to an exotic metal with strong electronic correlation. The molecules are weakly bonded to construct organic semiconductor crystals, so that the clean and coherent electronic systems can be formed with relatively small band width (~ 0.5 eV) and on-site Coulomb energy larger than the band width [1,2]. Employing a molecule composed of an elongated π-electron core and σ-electron units connected to both edges of the core, a quantum well is formed as the result of two-dimensional crystallization. We developed a method of continuous crystallization from solution using a meniscus to grow a-few monolayer crystal films to the size of 30 cm [3,4].
Among many examples of the quantum-well molecules, we have been mostly studying the system of double-molecular-layer alkyl-naftobenzodithiophene (Cn-DNBDT) crystals [2]. Upon doping with an electric double-layer gate of an ionic liquid, the 2D crystal layer of the π-electron core is well protected from invasion of the anions, so that high carrier mobility exceeding 20 cm^2/Vs is preserved even with 1/4 holes per molecule, resulting in the first observation of metallic phase in organic semiconductors [5]. The mean-free path is more than 5 times as long as the molecular distance, satisfying the Ioffe-Regel condition of $k_F l$ larger than unity. The sheet conductance is well above conductance quantum of e^2/h at low temperatures. At the high carrier concentration about 10^{14} cm^{-2}, which approximately fills 1/8 of the HOMO band, the carrier density estimated by Hall coefficient drastically diminishes with decreasing temperature below ~100 K. Therefore, it is likely that pseudo-gap opens at the Fermi level because of the pronounced on-site Coulomb energy due to poor screening in the system of inherently small band width. The pseudo-gap phase further dominates with higher carrier concentration in the region of present experiment, so that mechanism of the exotic correlation effect is under investigation with even higher carrier concentration or by employing other molecular compounds.

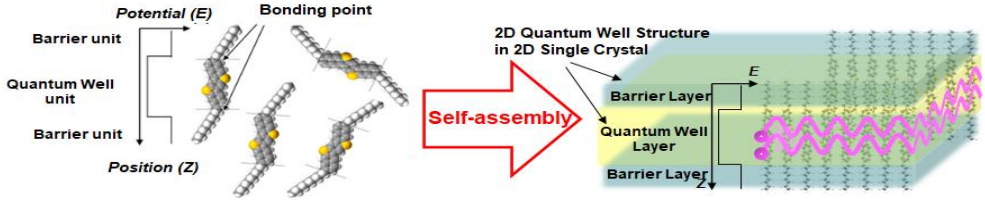

Fig. 1. Self-organized assembly of quantum-well molecules.

References

[1] J. Takeya et al., Jap. J. Appl. Phys. 44, L1393 (2005); Phys. Rev. Lett. 98, 196804 (2007).
[2] C. Mitsui, T. Okamoto, J. Takeya et al., Adv. Mater. 26, 4514 (2014).
[3] A. Yamamura, S. Watanabe, J. Takeya et al., Sci. Adv. 4, eaao5758 (2018).
[4] T. Sawada, S. Watanabe, J. Takeya et al., Nat. Commun. 11, 4839 (2020).
[5] N. Kasuya, S. Watanabe, J. Takeya et.al., Nature Mater. 20, 1401 (2021).