Two-dimensional hole gas in single-crystal organic semiconductors by ionic liquid gating

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Organic semiconductors (OSCs) are van der Waals solids which are composed of single component pi-conjugated molecules. Therefore, OSCs are classified into wide-gap intrinsic semiconductors¹. Electronic phase transition under high carrier density has been extensively studied in the field of OSCs. Any electronic phase transition, however, has not been observed in OSCs.

Here, we demonstrate two-dimensional hole gas (2DHG) in layered single-crystal semiconductor C8-DNBDT using electric double layer transistors (EDLT) and success the observation of metallic states of C8-DNBDT². The present C8-DNBDT has linear octyl chain connected to pi-conjugated system, leading to separated structure in which charge carriers accumulated at the pi-conjugated system is protected from extrinsic potential disorder. In this work, high carrier density up to 10^{14} cm⁻², corresponding to 0.25 holes per one molecule, is accumulated at the single-crystal layer of C8-DNBDT, resulting in positive coefficient of *dR/dT* with lower sheet resistance than resistivity quantum h/e^2 (~ 25.8 kΩ).

The temperature dependence of Hall mobility which increases with sample cooling down to ⁴Helium temperature suggests quantum mechanically confined degenerate hole system is realized in the single-crystal layer of C8-DNBDT. Whereas Hall carrier density $(eR_{\rm H})^{-1}$ ($R_{\rm H}$: Hall coefficient) decreases as temperature decreases. It implies that hole-hole interaction affects its electronic properties, especially density of states at the Fermi level.

These transport properties of layered single crystal of C8-DNBDT represents the formation of 2DHG with relatively strong hole-hole interaction.

[1] S. Fratini, et al., Nat. Mater. 19, 491 (2020). [2] N. Kasuya et al., Nat. Mater. 20, 1401 (2021).



Figure 1. (a), (b), (c) The molecular and crystal structure of C8-DNBDT. (d) The schematic image of an EDLT. (e) The temperature dependence of sheet resistivity with various carrier density.