On Controlling Charge Carrier Transport in Colloidal Quantum Dot Assemblies

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Colloidal quantum dot (QD) solids are solution-processable thin-films that exploit the quantum confinement properties of the constituent nanocrystals. The quantum confinement effect in these materials generates energy bandgap value variations by size and the quasi-atom-like discrete energy levels. These two essential properties of the QDs, are intriguing for applications in different kinds of energy converting or harvesting devices. Investigation of carrier transport and electronic properties of highly-coupled colloidal quantum dot (QD) assemblies is vital for utilizing these classes of materials in diverse emerging practical applications, which are still dominated by exploiting the materials optical properties. Furthermore, the diverse combinations of QD compounds, ligands/coupler substances, assembly orders, and the emerging properties due to their surface effect made the electronic transport study become non-trivial. Field-effect transistor (FET) offers one of the best platforms to characterize the charge-carrier transport in QD assemblies. Nevertheless, it is an interfacial device that may be surface-sensitive and mostly concern about carrier transport in the planar direction. Meticulous scrutiny to separate the intrinsic characteristics of the QD assemblies from the other factors is vital.

Here we establish a framework to understand the charge carrier transport of various QD assemblies acquired from FET measurements. The merit of FET is its capability to allow us to modify the charge carrier density in the assembly by field-induced doping. It strongly depends on how the coupling of the gate system to the probed materials that act as the FET channels. The QD assemblies' surface quality varies by different assembly methods, existing facet conditions, assembly orders, etc. Therefore, they become factors in comparing the use of solid gate and conformational gate, which are electrolyte gate (including ionic liquid gate) or top-deposited polymer gate. Besides the formed electric field by the gating, the dielectric coefficient of the QD, which is influenced by the quantum confinement effect, may also play a role in deciding how thick the charge accumulation layer is. The influence of the doping-like effect in the FET measurement outcomes, both in transfer characteristics and carrier injection characteristics, will be discoursed. Furthermore, the observations of band-filling effect at high carrier density accumulation in several Pb-chalcogenide systems will be reviewed and rethinking the occurrence of band half-filling as the gauge to determine many other parameters related to carrier concentration, mobility, and traps. Ref. [1] S. Z. Bisri, et al. *Adv. Mater.* 25, 4309 (2013); 26, 5639 (2014); [2] L. Liu, et al. Nanoscale 11, 20467 (2019); [3] R.D. Septianto, et al. *NPG Asia Materials* 12, 33 (2020); [4] R. Miranti, et al. *ACS Nano* 14, 3242 (2020). We acknowledge the contributions of L. Liu and D. Shin (formerly U. Tokyo) for some of the data.

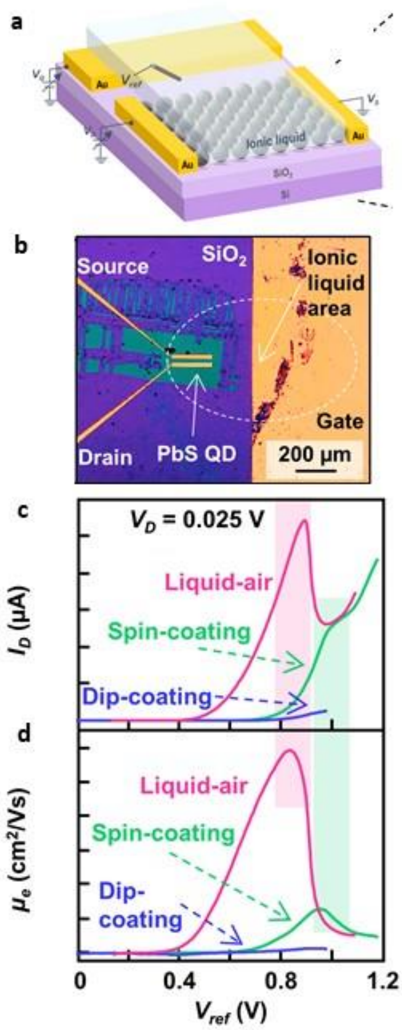

Fig. 1. (a) Schematic and **(b)** micrograph of electric-double-layer transistors (EDLT) of PbS QDs. **(c)** The comparison of the I_D-V_{REF} transfer characteristics of the PbS QD micro-EDLTs fabricated using different assembly methods. **(d)** The comparison of gate-dependent electron mobility of the devices.

