Accessing the Quantum Confinement Energy Levels in High Performance PbTe Colloidal Quantum Dot Transistors

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Colloidal quantum dots (CQD) assemblies emerge as a new type of hybrid solid thin films that exploit the size-dependent quantum confinement properties of the individual QDs: energy bandgap variations and formation of discrete energy sub-bands. Among lead chalcogenide (PbX) CQDs, lead telluride (PbTe) is the least explored, while it is prospective as materials of choice for highly-efficient size-tunable-range mid-infrared emitters and photodetectors, as well as high performance thermoelectrics. Field-effect transistors of PbTe CQD has only been demonstrated using hydrazine to replace the capping ligands of the QDs, which is unstable.[1] An effort to utilize thiocyanate ligands to crosslink the QDs was too challenging due to the slow rate of the ligands exchange.[2] These hampers the utilization of PbTe QDs for applications that requires adequate charge carrier transport.

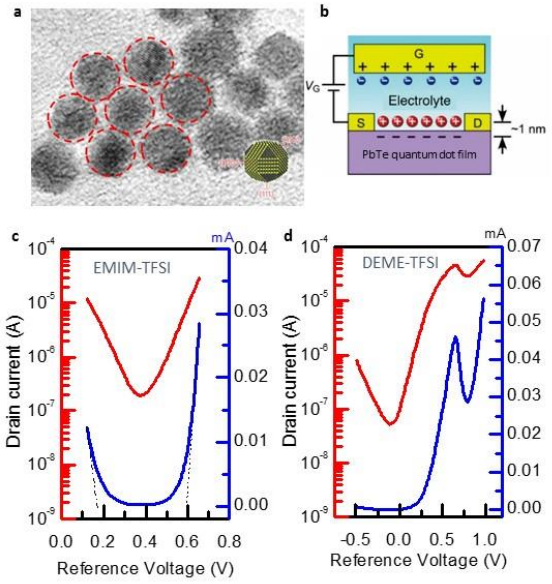

Figure 1 (a) TEM micrograph of PbTe CQD assembly crosslinked 3-MPA ligands. **(b)** Schematic of the EDLFET of PbTe CQD with EDT. **(c)** I_D-V_G characteristics of EDLFETs of PbTe CQDs that used EMIM-TFSI IL; and **(d)** DEME-TFSI IL. The applied V_D was 100 mV.

Here we demonstrate high performance FETs of PbTe colloidal QD assemblies that combines high carrier mobility and preservation of quantum confinement effect. Instead of using volatile hydrazine or thiocyanate, we exploit the developed knowledge from the studies of charge carrier transport of PbS CQDs. 3-mercaptopropionic acid (3-MPA) and ethanedithiol (EDT) were utilized to shorten the inter-QD distance and to crosslink the PbTe QDs to form multidomain hexagonal assemblies.[3] Electric-double-layer (EDL) gating using ionic liquid is employed since it can accumulate very large carrier density. It enables the filling of carrier traps that would enhance the charge carrier transport in the assemblies.[3] Ambipolar FETs with high carrier mobility are demonstrated with holes and electrons mobility can reach values in the order of >1 cm^2/V.s and >10 cm^2/V.s, respectively. Because of almost complete trap filling by the ionic liquid gating and the observation of ambipolar characteristics, we can utilize this FETs to probe the bandgap of the QD assembly from the threshold of both holes and electrons accumulation. In addition, the usage of ionic liquid with wider electrochemical window enable us to access the higher discrete energy subbands of the quantum dots. This can be inferred from the negative differential transconductance behavior of the FETs. This reproduces the QD energy level probing capability at room temperature using EDLT, as what had been done for PbS QDs.[4] From this, we can identify the quantum capacitance of this QD FET from which we can extract altogether the carrier density and mobility. These results are vital for the utilization of PbTe CQD assemblies for thermoelectrics, interband photodetectors, and photovoltaic with multiple exciton generation capability, where preservation of quantum confinement is important.

Refs: [1] J. Urban, et. al. *Nature Mater.* 6, 115 (2007); [2] A. Fafarman, et al. *JACS* 133, 15743 (2011); [3] S.Z. Bisri, et al. *Adv. Mater.* **25**, 4309 (2013); [4] ibid. *Adv. Mater.* **26**, 5639 (2014)