Colloidal Quantum Dot Arrays for Thermoelectric Devices

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Nanomaterials is one of the most promising building blocks that may overcome the challenges to develop high performance thermoelectric materials.[1] One of the most important feature of colloidal quantum dots (QDs) in this respect is the formation of quasi-atomic discrete energy levels resulting from the quantum confinement effect. This feature lead to sharp density of states that beneficial for thermoelectric, if we are able to fill them. Lead chalcogenide, either PbS and PbTe QDs are among the most prospective materials sought for this direction, because of its natural abundance and its known superb thermoelectric properties in its bulk form, respectively. While so far the most common approach to utilize colloidal QDs for thermoelectricity is limited as material sources for hot-pressed nanocomposite pellets and relies much on metal intercalation to enhance its electrical conductivity,[2] exploitation of the preserved quantum confinement effect has yet to be explored.

In this presentation, we show the importance of charge-density control on PbS and PbTe CQD assemblies by field-induced doping utilizing electric-double-layer (EDL) gating. Through optimization of quantum dot size, interdot distance, as well as the utilized molecular crosslinkers, we can achieve an improved orders of the QD assemblies, in particular PbTe QDs that have been so far difficult to assembled to form adequate conducting channel. The accumulation of high carrier density by EDL gating is not only enhance the electronic conductivity of the QD assemblies,[3] but also allow us to probe their electronic band-gap by transport measurement,[4] as well as accessing the preserved discrete energy levels. This is because the relatively narrow bandgap of both PbS and PbTe QDs (< 1 eV) than the limit of the Fermi level shift that can be governed by the EDLFET. The preservation of the discrete energy levels, indicated by the sharp electronic density of states, despite the large scale array of the assembly would enable us to expect the observation of high Seebeck coefficient when we access them. There is strong indication that we obtain high Seebeck coefficient once we access this discrete energy levels of these two QD system. Furthermore, the optimized high electrical conductivity that are achieved by these system can produce high power factor value from this material system. This provide new approach to develop efficient thermoelectric materials by controlling the appropriate carrier density required to dope quantum dot superlattice.

Refs: [1] J. Urban, *Nature Nanotech.* 10, 997 (2015); [2] M. Ibanez, et al. *Nature Comm.* **7**, 10766 (2016); [3] S.Z. Bisri, et al. *Adv. Mater.* **26**, 5639 (2014); [4] S.Z. Bisri, et al. *Adv. Mater.* **25**, 4309 (2013).

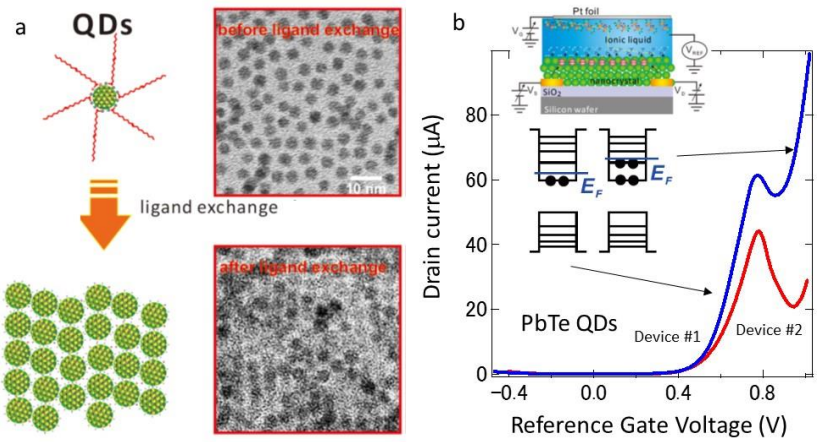

Figure 1 (a) Schematic diagram on the assembly of PbS colloidal quantum dot to form superlattice assembly by crosslinking them with shorter ligands and their respective transmission electron micrograph. (b) The I_D-V_G transfer characteristics of PbTe colloidal quantum dots gated using ionic liquid demonstrating negative transconductance that indicates the filling of the discrete energy levels. The same approach can be performed while measuring thermoelectric properties.