

Enhancing Electrical Conductivity in Lead Chalcogenide Colloidal Quantum Dot Assemblies via Remote Molecular Doping

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Assemblies of colloidal semiconductor quantum dots (QDs) emerge as solid thin films that exploit the size-dependent quantum confinement properties and the wet chemical methods, vital for the development of the emerging solution-processable electronics and optoelectronics technologies. The ability to control the carrier transport in the QD assemblies is fundamental for altering its electronic and optical properties based on the desired applications. For many of these device applications, some certain doping levels are required, in particular, to enhance the conductivity. In particular, for narrow-bandgap PbX (X = S, Se, Te) QD assemblies, greater control in increasing the conductivity either to the hole side or to the electron side are necessary for many applications where their intrinsic ambipolar characteristics should be suppressed. So far, the carrier conductivity in the QD assemblies is still very low, which partially stem from the low carrier mobility due to the isolated nature of the individual QD among the assemblies, in addition to the tendency to have large carrier trap sites due to the surface dangling bonds. There have been many efforts to reduce the distance between the QDs to enhance the coupling between them, while keep separating them by optimizing the use of molecular ligands. However, from the other viewpoint, the spaces between the QDs that “suppress” the coupling between them might provide another way to enhance the conductivity by allowing some additional dopants to be inserted into these spaces.

Here we show the demonstration of the enhancement of charge carrier conductivity in various assemblies of lead chalcogenide QDs by remote doping utilizing different types of organic molecules that can donate charge carriers into the QDs. These molecular dopants can be in the vicinity of the QDs or fill the void spaces in between the QDs. Several different types of charge transfer molecules are used to dope, via solution process, either PbS and PbTe QD assemblies that have been crosslinked by molecular ligands. The additions of these molecular ligands can enhance the charge carrier conductivity up to three orders of magnitudes, reaching a value close to the order of S/cm. While doping the colloidal QDs using these charge transfer molecular dopants, which some of them are widely used for organic electronics, seem straightforward, several complex parameters are stemming from the fundamentals of the QD system itself. Surface-related properties may alter some concept of doping. As a consequence, there are several counter-intuitive results, in which the use of some dopant molecules known to make materials to become n-type could result in enhanced p-type conductivity.

Refs.: [1] A. Stavrinadis, G. Kostantatos, *ChemPhysChem* 17, 632 (2016), [3] A. R. Kirmani, et al. *ACS Energy Lett.* 1, 922 (2016). [3] S.Z. Bisri, et al. *Patent pending*. Supports from M. Ibanez, M. Kovalenko (ETH Zurich) and Cambridge Display Technology, UK are acknowledged.

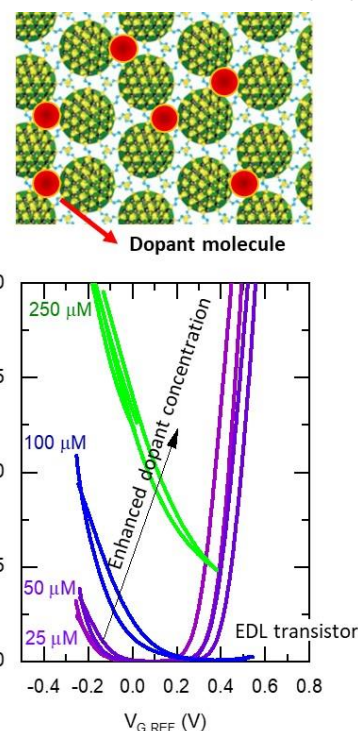


Figure 1. (top) Schematics of crosslinked PbX QD assemblies with molecular dopants fill the void spaces allowing them to donate charge carriers. (bottom) Comparison of I_D - V_G transfer characteristics of electric double-layer transistors of crosslinked PbTe QD assemblies with various concentrations of diluted molecular dopants.