Graphene – CdSe/ZnS quantum dots conjugated systems: charge transfer phenomena and their applications

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Graphene possesses unique physical properties, due to its specific energy bands configuration, substantially different from that of materials traditionally employed in solid-state electronics. Among the variety of remarkable properties, high transparency in the visible-light range and low resistivity of graphene sheets are the most attractive ones for optoelectronic applications.

Zero-dimensional colloidal semiconductor nanocrystals, known as quantum dots (QDs), attract immense attention in the field of photonics due to their size-dependent tunable optoelectronic properties. Their interaction with conductive surfaces is often reported to provide energy transfer instead of the charge transfer mechanism under presence of optical excitation. In this work, we demonstrate the role of single layer graphene (SLG) as an efficient charge transfer agent when conjugated with CdSe/ZnS core/shell quantum dots.

We first demonstrate quenching of QDs photoluminescence (PL) on the graphene surface. Firgure 1a shows single-layer graphene flake exfoliated on a glass substrate followed by dispersion of QDs. Figure 1b reveals PL image of the same area obtained via confocal microscope setup. A PL quenching of ~10-15 is observed in correspondence of the SLG area, when compared to the SLG-free areas covered with QDs. Figure 1c confirms the presence of QDs on graphene area, however, at magnified intensity scale.

We then characterize back-gated SLG field-effect transistors (FET) electrically during the QD excitation in order to investigate the nature of the interaction between SLG and the QDs. The transfer characteristics I_{ds} vs. V_g of back-gated SLG field-effect transistors (FET) are shown in figure 2. The pristine SLG FET has its neutrality point (V_{NP}) at ~ 0V. The same SLG FET is then sensitized by spin casting the QD solution. The transfer characteristics of the device are re-measured during excitation with laser of appropriate wavelength. A shift of the neutrality voltage V_{NP} from –1.9 V towards more negative voltages is observed (V_{NP} = –3.4 V), indicating that the electron accumulation in SLG is magnified by the interaction with the charge released by the optically excited QDs. The band alignment between SLG and the QDs allows electrons to be transferred by tunneling through the thin ZnS shell (figure 2b). The two conduction states (corresponding to the laser ON/OFF states) can be cycled reproducibly.

Additionally, we demonstrate an efficient electron injection from graphene into CdSeZn or CdSe/ZnS layered nanocrystalline structures and the operation of the corresponding graphene-QDs light emiting diodes (LED). Typical structure of graphene-QDs LED device is shown in figure 3. A sandwich fabricated on top of thin gold film (anode) consists of hole transport layer PEDOT:PSS, QDs layer and the single graphene layer, acting as cathode. In order to fabricate large-scale devices with active area of 1x1 mm^2 we utilize graphene layers synthesized via chemical vapor deposition (CVD) techniques. Graphene films are then transferred to target sandwich structures by traditional PMMA transfer process. Our devices reveal rather low threshold voltage of ~ 4.2 V and electroluminescence (EL) intensities up to ~300 cd/m^2. We demonstrate that the latter parameter can be increased dramatically by adjusting the work-function of graphene electrode via molecular doping.

References

[1] Klekachev, A.V., et al., Electron accumulation in graphene by interaction with optically excited quantum dots. Physica E-Low-Dimensional Systems & Nanostructures, 2011. **43**(5): p. 1046-1049.

Figures

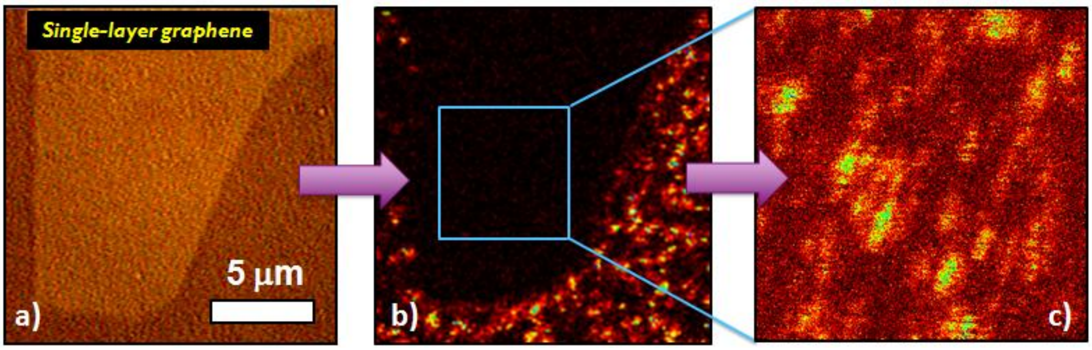

Figure.1: (a) Optical image of a graphene flake exfoliated on a glass cover slip. (b) Photoluminescence image of the same graphene flake after treatment with QDs. (c) Photoluminescence of individual QDs on the graphene in the area, selected by blue rectangle under different intensity scale.

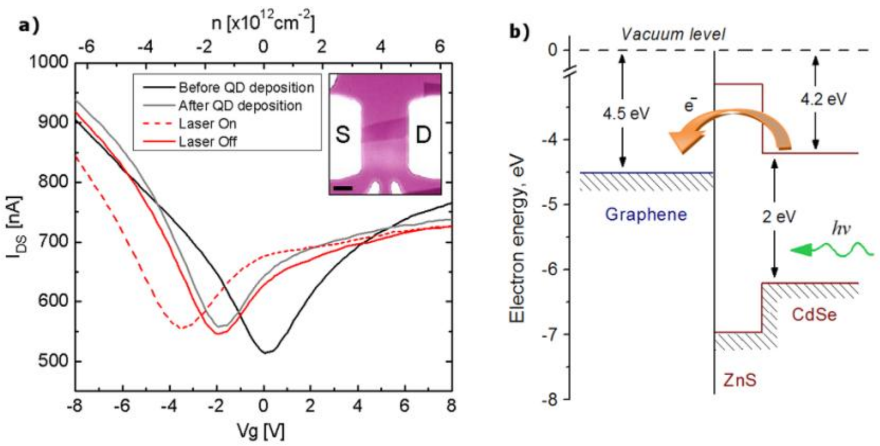

Figure.2: (a) Transfer characteristics of a SLG FET in pristine conditions, after QD deposition, and during QD excitation by a laser light. The inset shows an optical microscope image of the device (scale bar: 2 µm). (b) Energy level diagram at the interface of the graphene-QD system under study

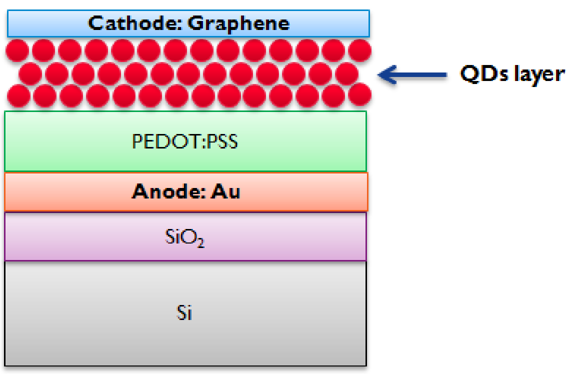

Figure.3: Schematic representation of graphene-QDs light emitting diode.