Electronic Supplementary Information (ESI)

Hexamethyldisilazane-mediated, full-solution-processed inverted quantum dot-light-emitting diodes

Yan Fu,[a‡] Daekyoung Kim,[b‡] Hyoungseok Moon,[a] Heesun Yang*[c] and Heeyeop Chae*[ab]

[a] School of Chemical Engineering, Sungkyunkwan University (SKKU), Suwon, 16419, Republic of Korea

[b] Sungkyunkwan Advanced Institute of Nanotechnology, Sungkyunkwan University (SKKU), Suwon, 16419, Republic of Korea

[c] Department of Materials Science and Engineering, Hongik University, Seoul 04066, Republic of Korea

Email: hchae@skku.edu (H. Chae), hyang@hongik.ac.kr (H. Yang)

Preparation of green CdSe@ZnS/ZnS QDs and ZnO NPs

Green-emitting multishelled CdSe@ZnS/ZnS QDs were synthesized according to the method reported elsewhere[1] with a minor modification. In brief, the cationic precursors of 0.14 mmol of Cd acetate and 3.41 mmol of ZnO with 7 ml of oleic acid (OA) were placed in a three-neck flask, degassed, N_2-purged and dissolved at 150°C for 30 min, followed by the injection of 15 ml of 1-octadecene (ODE) and further heating to 300˚C. Separately, the anionic stock solution was prepared by dissolving 3 mmol of Se and 3 mmol of S in 3 ml of trioctylphosphine (TOP) at 280°C, and 2.2 ml of this anionic stock solution was swiftly injected into the above hot cationic mixture and the reaction was allowed for 10 min for the growth of composition-gradient CdSe@ZnS QDs. Subsequently, a S-ODE solution, prepared by dissolving 1.6 mmol of S in 2.4 ml of ODE, was injected into the reactor at 310°C. After 12 min of reaction, a Zn-oleate solution, prepared by dissolving 2.86 mmol of Zn acetate dihydrate in 4 ml of OA and 1 ml of ODE, was introduced into the reaction. Finally, a S-TOP solution, comprising 10 mmol of S in 5 ml of TOP, was added slowly at a rate of 0.5 ml/min, followed by the reaction at 270˚C for 20 min to form ZnS outer shell. As-synthesized QDs were repeatedly purified with a solvent mixture of hexane and ethanol by centrifugation. The purified QDs were dispersed in a solvent mixture of octane/hexane (a volume ratio of 1/2) and their concentration was tuned by setting the optical density (OD) to 1.5 at 512 nm (excitonic absorption wavelength) for the following use for EML solution-deposition.

Colloidal ZnO NPs were prepared based on the earlier solution-precipitation protocol.[2,3] For a typical preparation of ZnO NP dispersion, 10 ml of 10 mM tetramethylammonium hydroxide (TMAH) solution in ethanol, was dropped into 40 ml of 10 mM Zn solution (zinc acetate dihydrate in dimethyl sulfoxide (DMSO)) for 15 min and this reaction was maintained for 1 h at 5°C. ZnO NPs formed thus were precipitated with excess acetone, collected by centrifugation, and finally dispersed in ethanol.

Full-solution-processed fabrication of inverted QLEDs

Indium tin oxide (ITO)-patterned glass substrate was cleaned sequentially with deionized (DI) water, acetone and isopropanol with a sonication and then UV-ozone-treated for 15 min. ZnO NP ETL with a thickness of 75 nm was generated on ITO substrate by spin-casting colloidal ZnO NP ethanol dispersion with a concentration of ~100 mg/ml at 1500 rpm for 30 s and annealed at 180°C for 20 min. Then the above QD dispersion in a mixed solvent of octane/hexane was mixed with 0.857 ml of HMDS and the resulting HMDS-blended QD dispersion was spin-casted at 3000 rpm for 30 s on top of ETL, forming 33 nm-thick HMDS-QD EML. EML of HMDS-QDs was then annealed at different temperatures of 130 and 160°C. And the subsequent 17 nm-thick HTL was generating by spin-casting poly-TPD dissolved in chlorobenzene with a concentration of 5 mg/ml at 3000 rpm for 30 s and annealed at 140°C for 20 min. A 10 nm-thick MoO_x HIL was formed sequentially by dissolving phosphomolybdic acid hydrate $(MoO_3)_{12}\cdot H_3PO_4\cdot(H_2O)_x$ (PMAH) in acetonitrile with a concentration of 10 mg/ml, spin-casting this precursor solution on poly-TPD HTL at 3000 rpm for 30 s and annealing it at 120°C for 10 min. Fabrication of multilayered inverted QLEDs were completed by thermally evaporating 100 nm-thick Al anode in high vacuum at a deposition rate of ~5 Å/s.

Characterizations

Absorption and photoluminescence (PL) spectra of QDs in both forms of dispersion and film were recorded with a UV-Vis spectrometer (JASCO, V-650) and a fluorescence spectrometer (Lumina™), respectively. The quantum yield (QY) of QDs was determined with a reference organic dye of rhodamine 6G (QY of ~95%) in ethanol with an identical OD of ~0.05 at 445 nm. To further confirm the above relative QY value, the absolute QY of QDs was assessed in an integrating sphere with an absolute quantum yield measurement system (C9920-02, Hamamatsu). Surface roughness of QD film was measured with an atomic force microscopy (AFM, SPA-300HV, Seiko). Particle and cross-sectional transmission electron microscopic (TEM) images of QDs and multilayered inverted QLEDs, respectively, were obtained with a JEM ARM 200F (JEOL) operating at 300 kV. All luminance−current density−voltage measurements, performed under ambient conditions (25°C, RH 50%), of the devices with an individual active area of 2×2 mm^2 were conducted with a source measure unit (2400, Keithley Instruments, Inc.) and a luminance meter (CS-2000, Konica Minolta Sensing, Inc.).

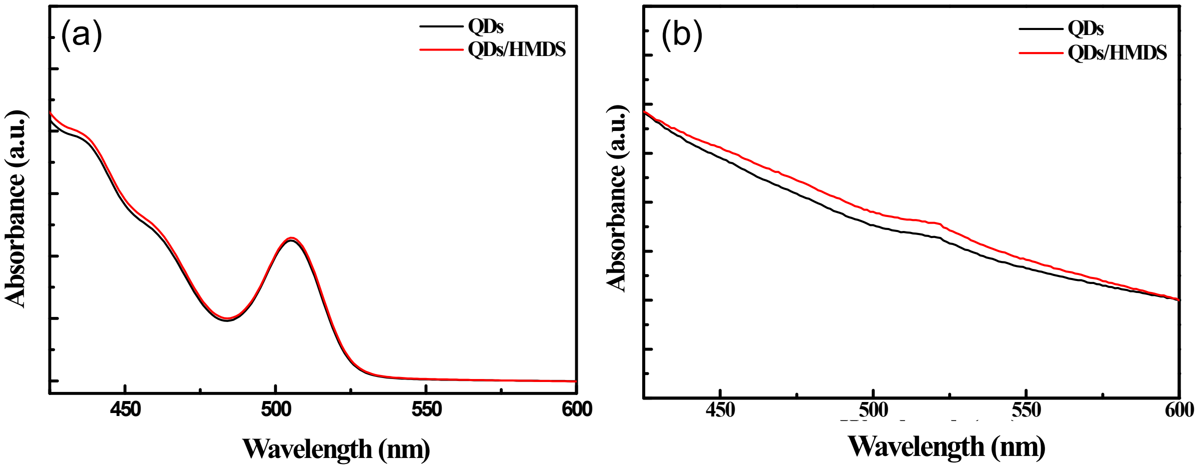

Fig. S1 Absorption spectra of green QDs without *versus* with HMDS additive in (a) solution and (b) film state.

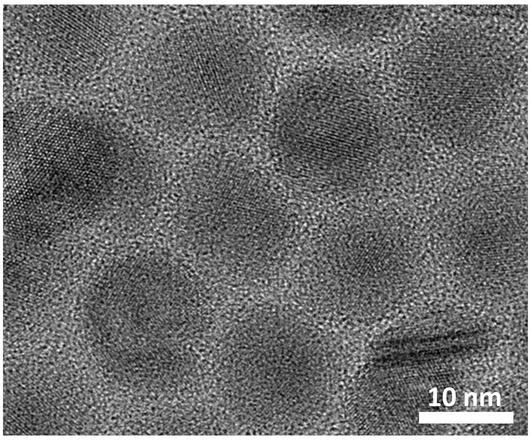

Fig. S2 TEM image of multishelled CdSe@ZnS/ZnS QDs with an average size of 13.5 nm.

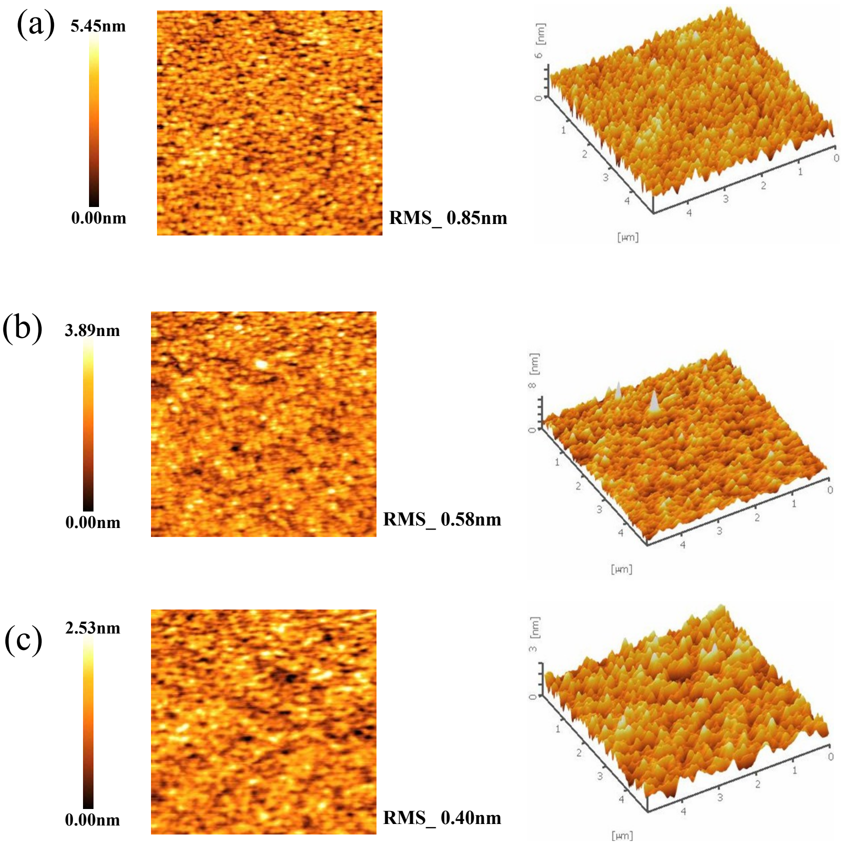

Fig. S3 AFM results of height (left) and three-dimensional images (right) of (a) 130°C-annealed pristine QD layer, HMDS-QD layers annealed at (b) 130 and (c) 160°C.

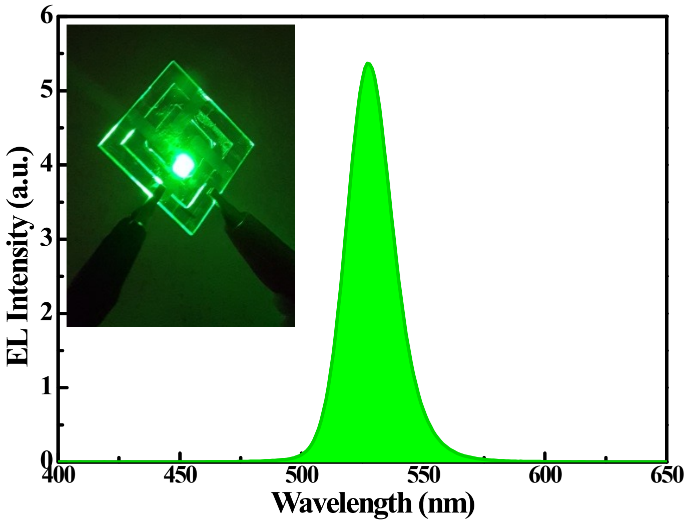

Fig. S4 A representative EL spectrum and EL image (inset) of HMDS-mediated inverted QLED collected at an operating voltage of 9 V.

References

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3. K. H. Lee, J. H. Lee, W. S. Song, H. Ko, C. Lee, J. Lee and H. Yang, *ACS Nano*, 2013, **7**, 7295−7302.