High-Performance Inverted Red InP Quantum Dot Light-Emitting Diodes with a New Electron Transport Layer

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ABSTRACT

Here we report high-performance inverted red InP QLEDs using a new KHU-ETL as an interlayer. Our optimized device with QD:DBTA(1wt%) showed a maximum EQE of 11.6% and a lifetime (LT_{50}) of 3800 hrs at 1000 cd/m^2. This enhancement is due to improved charge balance and also suppressed interfacial exciton quenching.

1 Introduction

Quantum dot light-emitting diodes (QLEDs) have gained tremendous interest in research due to their attractive properties such as high color purity, wide color gamut, high transparency, and simple fabrication process. These attractive properties make QLEDs an excellent candidate for next-generation display and solid-state lighting applications.[1] Recently, many researchers have focused on environment-friendly cadmium-free indium phosphide (InP) QDs. Won et. al achieved a maximum external quantum efficiency (EQE) of 21.4% with InP QDs after proper ligand exchange.[2] However, the device performance of cadmium-free InP QLEDs is inferior compared to cadmium-based QLEDs. The poor device performance of InP QLEDs is mainly due to electron and hole injection imbalance. Normally, ZnO is commonly used as an electron transport layer (ETL) because of its excellent properties such as high mobility, suitable energy level, and simple solution process. Due to high mobility of ZnO ETL compare to organic hole transport layer (HTLs), creates charge imbalance in the QD emissive layer which reduces the device performance. Therefore, charge balance plays a crucial role in improving the device performance of QLEDs. Peng. et. al utilized a thin insulating poly (methyl methacrylate) (PMMA) layer between the QD and ZnO ETL and achieved an excellent device performance.[3] Wang et. al used polyethyleneimine (PEI) insulator to decrease the electron injection and suppress the exciton quenching. However, the device performance mainly depends on thin insulator thickness, which is sensitive to control, indicating a lack of reproducibility. Another effective approach is by doping metals such as magnesium (Mg), gallium (Ga), and Yttrium (Y), etc., in ZnO can tune the mobility and conduction band minimum (CBM) level. Among all, 15% Mg doped ZnO ($Zn_{0.85}Mg_{0.15}O$) ETL shows excellent device performance by achieving good charge balance. However, the $Zn_{0.85}Mg_{0.15}O$ ETL is unstable due to intrinsic physical properties.[4]

Here we report a high performance inverted red InP QLED using a slow mobility KHU-ETL as an interlayer. By insertion of KHU-ETL interlayer between ZnO and QD EML, the electron injection is reduced, which improves the charge balance in the device and also suppresses the interfacial exciton quenching due to higher CBM level of KHU-ETL. Further, we adopted QD:DBTA(1wt%) approach, HTL doping in the QD to improve the hole injection. As a result, our optimized device with QD:DBTA(1 wt%) showed a maximum EQE of 11.6% and a device lifetime of (LT_{50}) of 3800 hours at an initial luminance of 1000 cd/m^2.

2 Experiment

The indium tin oxide (ITO) substrates were sequentially cleaned with acetone, and isopropyl alcohol for 10 mins each in an ultrasonic bath and finally washed with deionized water. The ITO substrates were dried with N_2 gas and followed by UV-ozone treatment for 10 mins. Firstly, ZnO was spin-coated on ITO substrates at 3000 rpm for the 60s and annealed at 200 ^{0}C for 60 mins. Later KHU-ETL interlayer is coated over ZnO at 3000 rpm for 60 sec and annealed at 250 ^{0}C for 60 mins. After that, red InP QD (8 mg/ml in octane) was spin-coated at 2000 rpm for 30 sec on the KHU-ETL interlayer. Finally, stacked HTLs, HIL and aluminum anode were thermally evaporated under high pressure.

3 Results and Discussion

Here, we used red InP/ZnSe/ZnS QDs for the fabrication of QLEDs. Fig. 1 shows the absorbance and PL spectra of red InP QDs. Our InP QDs showed a maximum PL peak at 630 nm and a narrow full width at half maximum (FWHM) of 36 nm with an absolute PLQY of 90%.

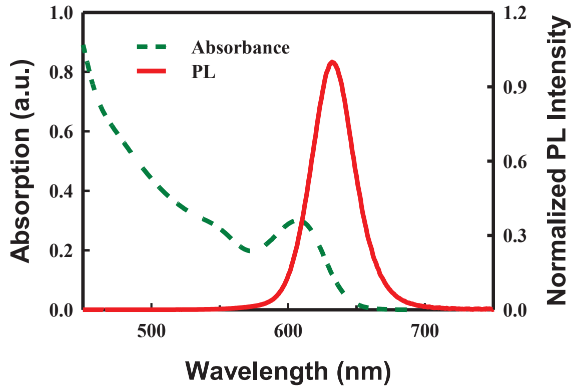

Fig. 1 Absorbance and PL spectra of red InP QDs.

To see the effect of KHU-ETL as an interlayer, we have fabricated an inverted QLED device with the following device structure.

Device structure: ITO/ZnO/KHU-ETL interlayer/red InP/ZnSe/ZnS QDs/DBTA/PCBBiF/HATCN/Al. We also fabricated a control device (without interlayer ZnO ETL) and well-known $Zn_{0.85}Mg_{0.15}O$ ETL for better comparison.

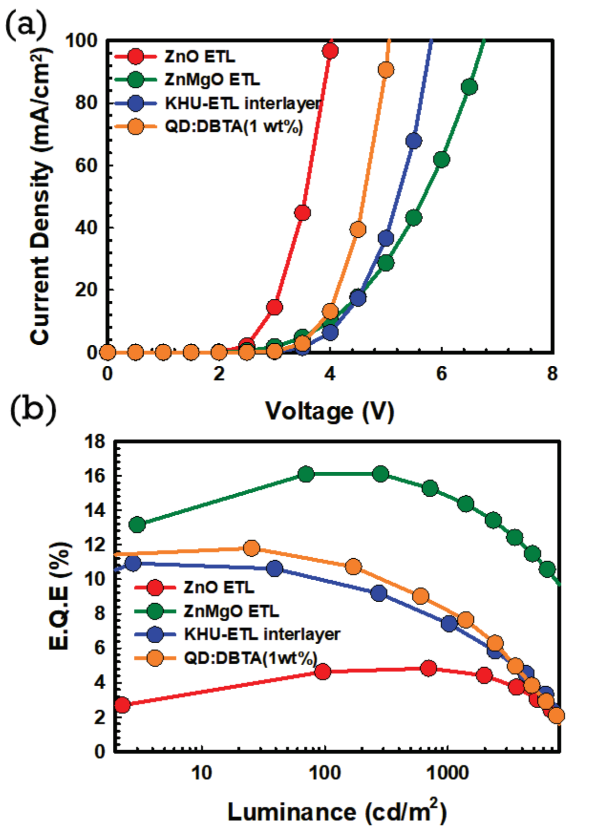

Fig. 2 (a) Current density versus voltage (b) EQE versus luminance characteristics of the fabricated red InP QLEDS devices with ZnO ETL, $Zn_{0.85}Mg_{0.15}O$ ETL, KHU-ETL interlayer and QD:DBTA(1wt%).

As shown in Fig. 2 (a), the device with ZnO ETL shows a faster current density. Whereas, the device with $Zn_{0.85}Mg_{0.15}O$ ETL shows a decreased current density. This is attributed due to the lower mobility of $Zn_{0.85}Mg_{0.15}O$ ETL and raised conduction band minimum (CBM). The device with a KHU-ETL interlayer shows decreased current density similar to $Zn_{0.85}Mg_{0.15}O$ ETL. Using KHU-ETL interlayer, the electron injection of ZnO ETL is reduced due to the higher CBM of KHU-ETL. Fig. 2 (b) shows the EQE versus luminance characteristics, the optimized QLED device with KHU-ETL interlayer shows a maximum external quantum efficiency (EQE) of 10.6%, which is 2.3-fold improved compared to the control device (EQE: 4.5%). The $Zn_{0.85}Mg_{0.15}O$ ETL device shows a maximum EQE of 16.1%, which is relatively higher than the interlayer device. This improvement in device performance is due to better charge balance and also reduced interfacial exciton quenching at ETL and QD interface. To further improve the charge balance with the KHU-ETL interlayer device, we adopted QD:DBTA(1 wt%) HTL doping in the emissive layer (EML) approach for efficient hole injection. The hole injection is relatively improved in the QLED device due to favorable energy levels. The optimized device showed a maximum EQE of 11.6%. Later, we have studied about the operating lifetimes of the fabricated QLED devices as shown in Fig. 3.

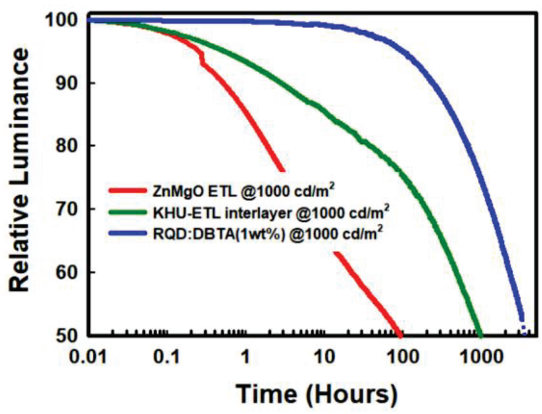

Fig. 3 Operating lifetimes of the fabricated red InP QLED devices with $Zn_{0.85}Mg_{0.15}O$ ETL, KHU-ETL interlayer and RQD:DBTA(1 wt%).

The inverted InP QLED device with $Zn_{0.85}Mg_{0.15}O$ ETL shows a half lifetime (LT_{50}) of 100 hours at an initial luminance of 1000 cd/m^2. By assuming acceleration factor n=1.8, the half lifetime at an initial luminance of 100 cd/m^2 is 6300 hours. Whereas with KHU-ETL interlayer device shows a half lifetime of 1000 hours at an initial luminance of 1000 cd/m^2 and 63,000 hours at an initial luminance of 100 cd/m^2. To know the reason behind the improvement in device lifetime, we performed an ETL stress degradation test on fabricated EOD devices. We have applied 15 mA/cm^2 stress is applied to check the stability of ETLs. After stress, the

$Zn_{0.85}Mg_{0.15}O$ ETL shows a decrease in operating voltage. This shows the unstable behavior of $Zn_{0.85}Mg_{0.15}O$ ETL to electron stress. The $Zn_{0.85}Mg_{0.15}O$ ETL is relatively sensitive to oxygen vacancies and hydroxyl bonds when stressed or stored due to intrinsic physical properties, which reduces the QLED device lifetime.[4] Whereas KHU-ETL interlayer EOD device shows no change in operating voltage up to several hours. This reveals that KHU-ETL is stable against electron stress. The half lifetime of QD:DBTA(1 wt%) doped device shows 3800 hours at an initial luminance of 1000 cd/m^2 and 239,400 hours at an initial luminance of 100 cd/m^2. The high efficiency and long device lifetime are mainly attributed due to reduction in electron injection and improvement in hole injection resulting in improved charge balance in the InP QLED device. Detailed results will be discussed at the presentation.

4 Conclusion

We report high performance inverted red InP QLEDs with a slow mobility KHU-ETL as interlayer. Our optimized device with QD: DBTA(1 wt%) showed a maximum EQE of 11.6% and half lifetime of 3800 hours at an initial luminance of 1000 cd/m^2. The enhancement in device efficiency and lifetime is mainly attributed due to improved charge balance and reduced interfacial exciton quenching at ETL and QD interface. We believe that our study will be useful to attain high performance QLEDs.

References

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