Highly Efficient Quantum Dot Light-emitting Diode Based on Properly Charge Balanced and Suppressed Interfacial Exciton Quenching Process

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ABSTRACT

We report a high efficiency inverted red indium phosphide (InP) based quantum dot light-emitting diode (QLED) by optimizing charge balance and suppressing interfacial exciton quenching process. Our optimized red InP-QLED using new deep HOMO level and high mobility hole transport layers and sol-gel ZnMgO showed external quantum efficiency of 21.8 % and current efficiency of 23.4 cd/A.

1. INTRODUCTION

A Light-emitting diode (LED) containing quantum dots (QDs) has emerged as the favorable technology for nextgeneration display applications due to high color purity, wide color gamut, and simple fabrication process. Recently, significant works have been done for the development of efficient indium phosphide (InP) QDs.¹ Therefore, the external quantum efficiencies (EQEs) of InP-QLEDs have been reached to ~20% (for red color).1 However, the efficiency and stability are still lower than the commercial requirement. Normally, the efficiency of InP-QLED is limited by the charge unbalance in the emissive QDs and low photoluminescence quantum yield (PLQY) of InP-QDs in the film state. The charge unbalance in QLED is linked to the charge injection barrier between the hole or electron transporting layer (ETL or HTL) and emissive QDs and the mobility of charge transporting layers.

Generally, inverted QLEDs are fabricated by using ZnMgO or ZnO nanoparticles (NPs) as an ETL and organic HTLs.² However, the conventional organic HTLs show low hole mobility than the inorganic ZnO or ZnMgO and also shows large charge injection barrier between the HTL and QDs, resulting in poor charge balance in the device.³ Also, the strong exciton quenching was observed at the interface of ZnO or ZnMgO NPs and InP-QDs due to the surface defects assisted non-radiative recombination process, resulting in lower device efficiency.³ Therefore, to make a high efficiency InP-QLEDs, proper charge balance into the emissive QDs and suppression of interfacial exciton quenching between the InP-QDs and ETL are required.

Here, we present a high-efficiency inverted red InP-QLED by optimizing a charge balance into emissive InP- QDs and suppressing the interfacial exciton quenching process. To improve charge balance into the emissive InP-QDs, new high mobility and deep HOMO level HTLs were used in a cascade structure. Also, the interfacial quenching process between ZnMgO and InP-QDs was suppressed by reducing the surface defects. Our inverted red InP-QLED with sol-gel ZnMgO and new high mobility and deep HOMO level HTLs exhibited a maximum EQE of 21.8%, which is the highest efficiency value reported for red InP-QLEDs. This result shows a significant improvement in the efficiency of the optimized device compared to the reference device.

2. EXPERIMENT

The indium tin oxide (ITO) patterned glass substrates were sequentially cleaned using acetone, isopropyl alcohol, and deionized water followed by UV-ozone treatment for 10 minutes. Firstly, sol-gel ZnMgO was spin-coated onto the ITO/glass substrates and annealed at 200 °C for 30 minutes. After that, red InP-QDs (10 mg/ml in octane) was spin-coated at 3000 rpm onto ZnMgO. Finally, new organic HTLs and metal cathode were deposited using a thermal evaporator under the high-vacuum evaporation chamber.

3. RESULTS AND DISCUSSION

Here, red InP/ZnSe/ZnS QDs were used for the fabrication of QLEDs. Our InP-QDs exhibited a maximum PL peak at 626 nm with a narrow FWHM (38 nm), PLQY of 90% in the solution state, and an exciton decay lifetime of 32.2 ns in the film state.

First, to suppress a non-radiative recombination process at the interface of ZnMgO NPs and InP-QDs, we used a properly optimized sol-gel ZnMgO layer due to its lower conductivity and fewer surface defects. The transient PL properties of the InP-QDs spin-coated on the sol-gel ZnMgO layer or ZnMgO NPs layer are shown in Fig 1a. The exciton decay lifetime of the InP-QD/ZnMgO NPs sample was reduced to 22.8 ns from 32.2 ns (InP-QDs) due to the higher surface defects. However, a sample with an optimized sol-gel ZnMgO layer exhibited an exciton decay lifetime of 25.9



Fig. 1 a) Transient PL of InP-QDs deposited on ZnMgO NPs and sol-gel ZnMgO. (b) Current density versus voltage characteristics of EODs and HODs.

ns. Also, the exciton lifetime of the InP-QDs was increased to 30.3 ns after using a surface treatment on the optimized sol-gel ZnMgO layer due to the suppressed non-radiative process. Moreover, we checked the effect of optimized sol-gel ZnMgO on the electron transport property by fabricating electron only devices (EODs). The EOD with an optimized sol-gel ZnMgO layer showed a significant reduction in the current density compared to ZnMgO NPs based device. These results show that the optimized solgel ZnMgO not only suppressed interfacial exciton quenching but also decreases the charge transfer process due to its lower conductivity than the ZnMgO NPs.

Normally, the efficient inverted QLEDs are fabricated with the thermally evaporated cascade organic HTLs, such as TCTA/TAPC or NPB/HATCN for fast transport of holes into the QDs.⁵ However, these HTLs have very low hole mobility and poor stability. Therefore, we designed new HTLs with high mobility and deep HOMO level (KHU-HTL1 and KHU-HTL2) than the reference materials. To check the effect of new HTLs on the hole transport properties, the hole only devices were fabricated with InP-QDs and new HTLs. As shown in Fig. 1b, the hole current density was significantly improved compared to the reference materials. Such high hole current density is attributed to the high hole mobility and deep HOMO level of new HTLs.



Fig. 2 The fabricated structure of inverted InP-QLED.

To confirm the influence of our optimized sol-gel ZnMgO layer with surface treatment and new HTLs on the electroluminescence performances, inverted red InP-QLEDs were fabricated with the structure shown in Fig. 2. Also, the reference device was fabricated with ZnMgO NPs and TCTA/TAPC HTLs for comparison of the device performances. The reference device with ZnMqO NPs and TCTA/TAPC HTLs showed lower driving voltage (4.3 V at 1000 cd/m²) than the optimized QLED (6.8 V) due to the higher conductivity of ZnMgO NPs. However, the optimized device exhibited an extremly high EQE of 21.80% and current efficiency of 23.46 cd/A with low-efficiency roll-off due to the proper change balance into InP-QDs and suppressed interfacial exciton guenching between sol-gel ZnMgO and InP-QDs (Fig. 3). These values are very higher than the reference device (7.50% and 7.31 cd/A). To the best of our knowledge, this is the highest efficiency reported for red InP-QLED. Also, our device showed CIE color coordinates of (0.68, 0.32), and maximum luminance of 23300 cd/m².



Fig. 3 EQE versus luminance of the red InP-QLED.

4. CONCLUSIONS

In summary, we demonstrate a high performance inverted red InP-QLED by optimizing charge balance into InP-QDs and suppressing interfacial exciton quenching between ZnMgO and InP-QDs. The charge balance into InP-QDs was improved by using surface treated sol-gel ZnMgO layer and new high mobility and deep HOMO level HTLs. Also, the interfacial exciton quenching process between ZnMgO and InP-QDs was suppressed by reducing surface defects and non-radiative recombination process. Our optimized red InP-QLED showed maximum EQE of 21.8% and current efficiency of 23.46 cd/A. To the best of our knowledge, this is the highest EQE reported for inverted red InP-QLED. We believe that our findings will be very useful for achieving high performances in the next-generation QD based displays.

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