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Studies of Electroluminescent Characteristics of Quantum Well Green Organic Light Emitting Diodes

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The electroluminescence (EL) characteristics are studied in organic light emitting diodes consisting of the emitting layer of tris(8-hydroxyquinolino)-aluminum (Alq_3) and the hole transporting layer of the type-II quantum well (QW) structure with 4,4',4''-tris(3-methylphenylphenylamino)triphenylamine (m-MTDATA)/4,4'-bis[N-(naphthyl)-N-phenyl-amino]biphenyl (α -NPD) as the quantum well/quantum wall. The EL spectra show a blue shift, around 10 nm compared with the reference device without the QW structure, as the number and thickness of quantum well and the current density increase. The luminescence can be controlled by 25% with the number of quantum wells and 20% with the thickness of the quantum well. The improvement of luminescence and the blue shift in the EL spectrum can be explained by the confinement of holes in the QW and the shifting of emitting zone.

Keywords Emitting zone; exciton; OLED; quantum well structure

1. Introduction

The application of quantum physics for optoelectronic device technology has been of keen interest for researchers since long time. The use of superlattice and multilayer quantum well (MQW) structure gives interesting electronic and optical properties of organic semiconductor devices. The multiple quantum well structure can affect the charge transport and optical spectrum of the organic light emitting diodes (OLEDs) through the confinement of charges in the well and control of energy transfer, respectively. Both of these properties can be exploited for color tuning and lowering power consumption of OLEDs.

The performance of OLEDs (e.g., external quantum efficiency) depends on carrier injection and recombination as well as the balance of electrons and holes [1–5]. All these parameters of the OLED performance can be tuned by using the proper

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choice of quantum wells, either type-II quantum well or type-I quantum well [6–8]. Many quantum well structures like tris(8-hydroxyquinolino)-aluminum (Alq_3)/2-(4-biphenyl)-5(4-tert-butyl-phenyl)-1,3,4-oxadiazole (PBD), N,N'-diphenyl-N,N'-bis(3-methylphenyl)-[1,1'-biphenyl]-4,4'-diamine (TPD)/ Alq_3 , N,N'-diphenyl-N,N'-bis(1,1-biphenyl)-4,4'-diamine (NPB)/2,3,6,7-tetramethyl-9,10-dinaphthyl-anthracene (TMADN), NPB/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP), NPB/copper phthalocyanine (CuPC), NPB/rubrene and m-MTDATA/ α -NPD have been reported [9–13]. The luminous efficiency and power efficiency increase in all the structures due to increase of electron-hole balance in the device. However, the mechanism of electroluminescence processes in the organic quantum well are still not clear and need more discussion.

The present work is an attempt to understand the electroluminescence (EL) characteristics in the type-II quantum well structure of m-MTDATA/ α -NPD in green OLEDs.

2. Experimental

The OLEDs with the organic quantum well structure were prepared by thermal evaporation of organic molecules onto ITO substrates which were ultrasonically cleaned with solvents. The thermal evaporation of organic materials and LiF/Al electrodes were conducted under the base pressure of 5×10^{-6} Torr without breaking the vacuum. The evaporation rate was monitored by a quartz-oscillator thickness monitor and maintained at $1 \sim 2$ Å/sec for organic materials and $2 \sim 3.5$ Å/sec for Al.

The QW device structure is ITO/ α -NPD:MoO₃ (x nm)/[m-MTDATA (y nm)/ α -NPD (5 nm)] _{n} /Alq₃ (50 nm)/LiF (0.5 nm)/Al (100 nm). Here, n ($=1, 2, \dots, 5$) is the number of quantum wells. The total thickness of the hole injection and transporting layer is fixed at 50 nm, *i.e.*, $x + n(y + 5) = 50$ nm in order to minimize the optical microcavity effect arising from the variation of the emitting zone relative to the electrodes [14]. The reference device structure is ITO/ α -NPD:MoO₃(10 nm)/ α -NPD (40 nm)/Alq₃(50 nm)/LiF (0.5 nm)/Al (100 nm). Since the device with $y = 4$ nm shows the best performance in the case of $n = 1$ compared with the reference device, we fabricated the multiple QW devices with the quantum well (m-MTDATA) and wall (α -NPD) thicknesses of 4 and 5 nm, respectively. We studied the device performance and EL spectrum shift with the variation of the number of quantum wells ($1 \leq n \leq 5$) as well as the quantum well width (y).

Current-voltage (I-V) characteristics were measured by using Keithley-236 source measurement unit, and luminance and external quantum efficiency (EQE) were calculated from the photocurrent measured with calibrated Si photo-diode (Hamamatsu S5227–1010BQ). Electroluminescence spectra were measured by using a monochromator (Acton ARC275) with PMT detector.

3. Results and Discussions

Figure 1 shows the current density – voltage characteristics for OLEDs with (a) increasing number of quantum wells ($n = 0 - 5$) and (b) with different quantum well (m-MTDATA) thickness ($y = 1 - 5$ nm) and the fixed quantum wall (α -NPD) thickness of 5 nm. Figure 1(a) shows that the current density decreases as the number of quantum well increases at the same operating voltage. This indicates the confinement of holes in the quantum well structure. Since the energy barrier for the hole injection is about 0.4 eV at the interface of m-MTDATA and α -NPD, the holes are

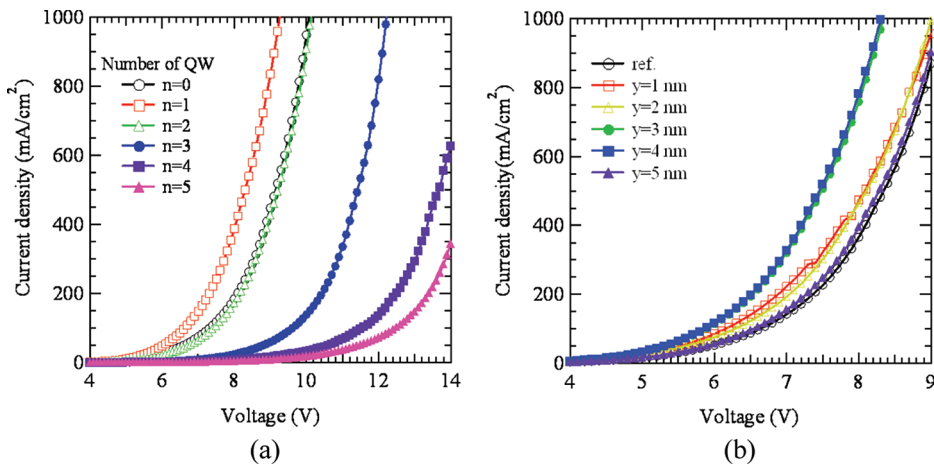


Figure 1. The current density – voltage characteristics for OLEDs with (a) increasing number of quantum wells ($n = 0 - 5$) and (b) with different quantum well (m -MTDATA) thickness ($\gamma = 1 - 5$ nm) and the fixed quantum well (α -NPD) thickness of 5 nm.

accumulated and confined in the quantum well structure and the holes transferred to the Alq_3 layer decrease with the number of quantum wells [13]. As a result, the balance of electron hole concentrations can be improved by the control of the quantum well structure, resulting in improved EL efficiency. Figure 1(b) shows that the current density at constant operating voltage is growing up to $\gamma = 4$ nm and it is almost same as the reference device at $\gamma = 5$ nm. The well thickness up to 4 nm behaves as the quantum well and above 4 nm it acts just like the hole transport layer (HTL) without the QW structure. The details of carrier confinement in the quantum well OLEDs of the same structure and the discussion on the schematic energy diagram can be found in our previous paper [13].

Figure 2 shows the luminance of the QW OLEDs as a function of (a) the number of quantum wells and (b) the quantum well thickness at the constant current density

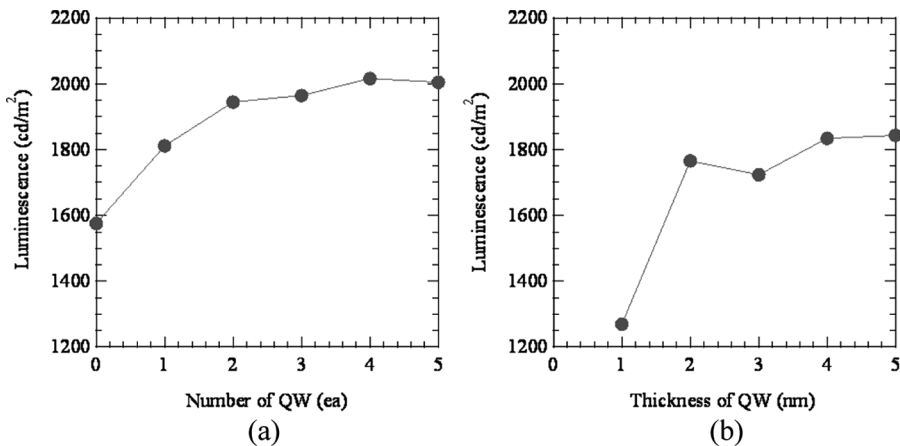


Figure 2. The luminance at the current density of $50 \text{ mA}/\text{cm}^2$ for (a) different number of quantum wells and (b) variation of quantum well thickness.

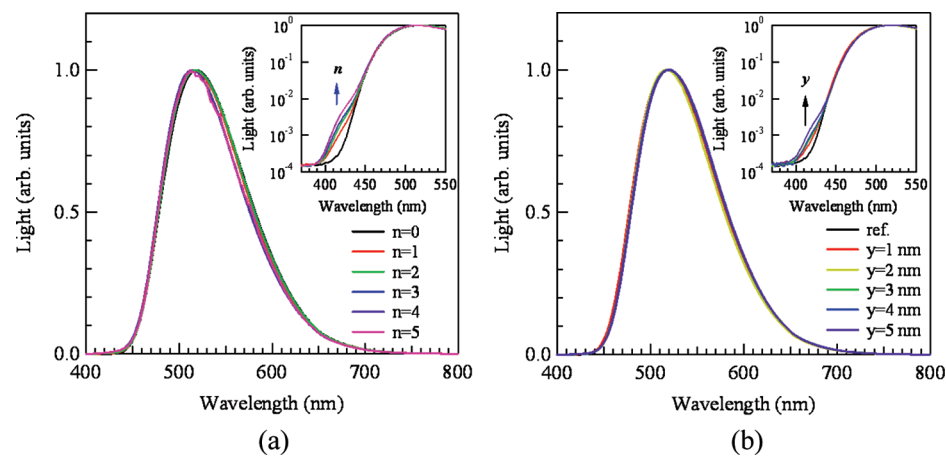


Figure 3. The EL spectra of OLEDs with increasing (a) the number of quantum wells ($n = 0 - 5$) and (b) the thickness of m -MTDATA ($y = 1 - 5$ nm) in a linear scale. In the inset of each figure we plot the blue spectral region in a logarithmic scale.

of 50 mA/cm^2 . The luminescence increases with increasing the number of quantum wells and it increases about 25% for the OLED with 5 quantum wells ($n = 5$). It can be also increased up to about 20% by increasing the well thicknesses up to $y \sim 4$ nm. The improvement of luminescence is due to the carrier confinement and balance of electron-hole in the device with the QW structure.

Figure 3 shows the EL spectra of OLEDs with increasing (a) the number of quantum wells ($n = 0 - 5$) and (b) the thickness of m -MTDATA ($y = 1 - 5$ nm) in

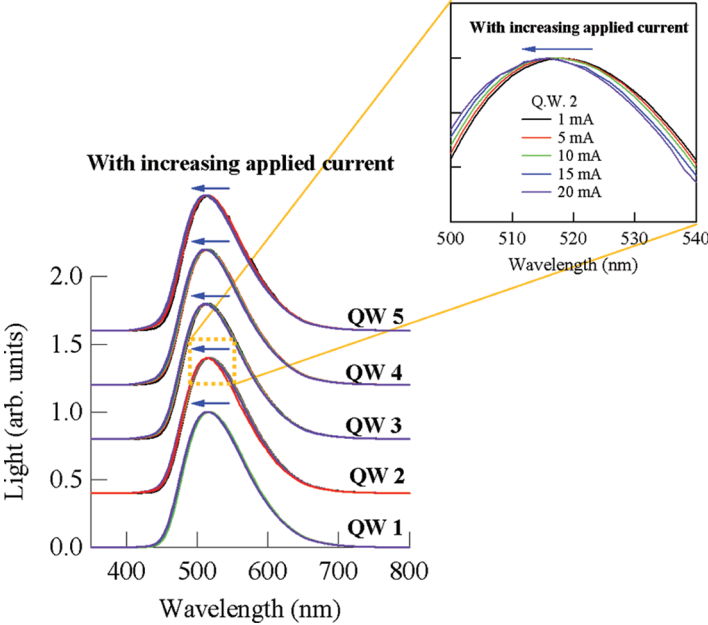


Figure 4. The variation of EL spectra of OLEDs with different number of quantum wells under various applied current densities.

a linear scale. In the inset of each figure we plot the blue spectral region in a logarithmic scale. The EL emission in the blue (400–450 nm) region arises from the α -NPD layer. This blue EL intensity increases as the number of quantum wells or the thickness of m -MTDATA increases. This behavior indicates the hole confinement at the QW structure as explained above in Figure 1. The accumulated holes at the m -MTDATA/ α -NPD interfaces can recombine with electrons entered from the Alq₃ layer through tunneling. Since the number of accumulated holes increases as the number of quantum wells increases, the blue EL intensity increases. Because of the additional blue emission from α -NPD and the shift of emission zone from the Alq₃ layer to the interface of Alq₃ and α -NPD due to the confinement of holes in the QW, the EL peak moves slightly toward the blue region with increasing the number of quantum wells at constant applied voltage.

Figure 4 shows the EL spectra for OLEDs with the number of QWs $n = 1 - 5$ for various current density. All EL spectra (QW $n = 1 \sim 5$) show a slight blue shift with the applied current. The spectral shift in the EL spectra for quantum well 2 is shown in a separate large graph window. The shift towards the interface due to the confinement of holes in the quantum well structure can be well understood by the EL spectra of QW OLEDs with the applied current.

4. Conclusions

The electroluminescence characteristics of multiple QW OLEDs were studied with different number of quantum wells and thickness of the quantum well. The QW structure can efficiently control the holes in the device and increases the electron-hole balance in the device. It results the increase of luminescence with the number of quantum wells and the thickness of the quantum well by 25% and 20%, respectively. The hole confinement in the quantum well structure causes the shift of the emitting zone in the device and results in a slight blue shift of the EL spectra.

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