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Luminance Efficiency Enhancement in Organic Light-Emitting Devices Utilizing 4,7-Diphenyl-1,10-phenanthroline/aluminum tris(8-hydroxyquinoline) Multiple Heterostructures Acting as an Electron Transport Layer

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The electrical and optical properties of organic light-emitting devices (OLEDs) with three periods of 4,7-diphenyl-1,10-phenanthroline (BPhen)/aluminum tris(8-hydroxyquinolate) (Alq₃) multiple heterostructures acting as an electron transport layer (ETL) were investigated. While the leakage current of OLEDs with multiple heterostructures was smaller than that of OLEDs without multiple heterostructures, the luminance efficiency was larger than that of OLEDs without multiple heterostructures. The BPhen layers in the multiple heterostructures blocked holes from the emitting layer (EML) to the ETL, and they enhanced the electron injection from the cathode to the EML, resulting in an increase in the luminance efficiency.

Keywords: organic light-emitting diode; multiple heterostructure; current density; luminance efficiency; electron transport layer; hole blocking

Introduction

Organic light-emitting devices (OLEDs) have become particularly attractive because of their potential applications in the fabrication of full-color flat-panel displays because they offer the unique advantages of fast response, low driving voltage, low power consumption, high color gamut, high contrast, and fast response [1–5]. OLEDs with multiple heterostructures have been introduced to obtain narrower spectral emissions, higher emission efficiencies, and tunable emission spectra [6–9]. Among the various methods for enhancement in the luminance efficiency of OLEDs, OLEDs utilizing a 4,7-diphenyl-1,10-phenanthroline (BPhen) layer with a higher electron mobility in comparison with a tris(8-hydroxyquinoline)aluminum (Alq₃) acting as an electron transport layer (ETL) have been reported [10]. Even though some works on the enhancement of the efficiencies of the OLEDs by using a BPhen ETL or a multiple heterostructure ETL have been suggested to improve the balance of holes and electrons in the emitting layer (EML) [11, 12], very few

studies concerning the OLEDs with multiple heterostructures consisting of a BPhen and an Alq₃ layers acting as an ETL have been reported.

This paper reports the luminance efficiency enhancement in OLEDs utilizing BPhen/Alq $_3$ multiple heterostructures acting as an ETL. The electrical and the optical properties of the OLEDs with BPhen/Alq $_3$ multiple heterostructures acting as an ETL were compared with those of the OLEDs with an Alq $_3$ single ETL or a BPhen:Alq $_3$ mixed ETL. Current density-voltage, luminance efficiency-current density, and electroluminescence (EL) measurements were carried out to investigate the electrical and the optical properties of OLEDs with a multiple heterostructure ETL.

Experimental Details

The sheet resistance and the thickness of the indium-tin-oxide (ITO) thin films coated on glass substrates used in this study were 15 Ω /square and 100 nm, respectively. The ITO coated substrates were cleaned by using acetone and methanol at 60°C for 15 min, and rinsed in de-ionized water thoroughly. The chemically cleaned ITO substrates were kept for 48 h in isopropyl alcohol. After the chemically cleaned ITO substrates had been dried by using N_2 gas with a purity of 99.9999%, the surfaces of the ITO substrates were treated with an oxygen plasma for 2 min at an O_2 pressure of approximately 2×10^{-2} Torr. The three kinds of OLEDs used in this study were deposited on ITO coated glass substrates by using organic thermal evaporation with effusion cells and shutters and consisted of the following structures from the bottom: an ITO anode electrode, an N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) (40 nm) hole transport layer (HTL)/an Alq₃ (30 nm) EML/three kinds of ETL (30 nm)/a lithium quinolate (2 nm) electron injection layer/an Al (100 nm) cathode electrode. The fabricated OLEDs contained three kinds of ETLs; an Alq₃ single layer (device I), a BPhen:Alq₃ mixed layer (device II), and three periods of BPhen/Alq₃ multiple heterostructures (device III). The electrical and the optical properties of device III with three periods of BPhen/Alq3 multiple heterostructure ETL were compared with those of devices I and II. Because the lowest unoccupied molecular orbital (LUMO) level of the BPhen layer is almost same as that of the Alq₃ layer, the electron injection barriers of devices I, II, and III are not significantly different from one another. The BPhen layer is used as an ETL or a hole blocking layer due to their higher electron mobility and lower highest occupied molecular orbital (HOMO) level. The thickness of the multiple heterostructure ETL for device III is the same as that of the ETL for other devices. Each thickness of the BPhen and the Alq₃ layers in the OLEDs with the multiple heterostructure ETL is 5 nm. The current density-voltage characteristics of the OLEDs were measured on a programmable electrometer with built-in current and voltage measurement units (SMU-236, Keithely). The luminance was measured by using a chromameter (CS-100A, Minolta), and the EL spectrum was measured by using a luminescence spectrometer (LS50B, Perkin-Elmer).

Results and Discussion

Figure 1 shows the schematic energy band diagrams for devices I, II, and III. The ETLs of devices I, II, and III consist of an Alq₃ layer, a BPhen:Alq₃ mixed layer, and three periods of BPhen/Alq₃ multiple heterostructures, respectively. Because the LUMO level of the BPhen layer is almost same as that of the Alq₃ layer, the electron injection barriers of devices I, II, and III are not significantly different from one another. The BPhen layer is used as an ETL or a hole blocking layer due to its higher electron mobility and lower

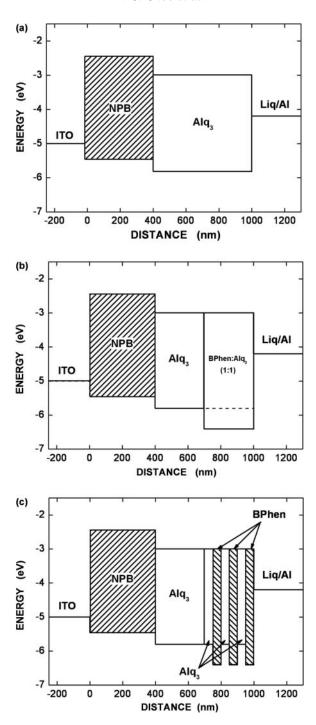


Figure 1. Schematic energy diagrams for devices (a) I, (b) II, and (c) III.

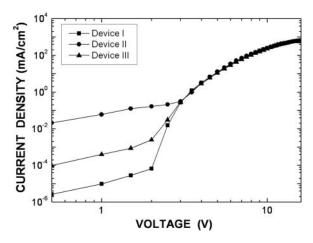


Figure 2. Current densities as functions of the applied voltage for devices I, II, and III.

HOMO level. The HOMO levels of the NPB, the Alq₃ and the BPhen layers are -5.5, -5.8 and -6.4 eV, respectively, and the corresponding LUMO levels are -2.4, -3.0, and -3.0 eV, respectively [13–15].

Figure 2 shows the current densities as functions of the applied voltage of devices I, II, and III. While the current densities of devices I, II, and III above 3 V are almost same, those below 3 V are significantly different from one another. While the current densities below 3 V are linearly proportional to the applied voltage, the corresponding leakage current densities do not significantly contribute to the emission. While the leakage current of device I with an Alq₃ ETL is the smallest value, that of device II with a BPhen:Alq₃ mixed layer is the largest value. The leakage current of device II with a mixed ETL increases in comparison with that of device I with an undoped ETL due to the coevaporation of BPhen and Alq₃ molecules during growth of a mixed ETL of device II. The leakage current of device II with a multiple heterostructure ETL at a low voltage range is larger than that of device I due to an increase of the internal electric field generated from the accumulated electrons at the Alq₃/BPhen heterointerface.

Figure 3 shows (a) the luminances as functions of the applied voltage and (b) the luminance efficiencies as functions of the current density of devices I, II, and III. While the luminance efficiency of device I rapidly decreases with increasing current density, those of devices II and III slightly decreases. The luminance and luminance efficiency of device III at an applied voltage of 10 V and at a current density of 200 mA/cm² significantly increase due to the increase of the electron injection through the BPhen/Alq3 multiple heterostructure ETL. The maximum luminance of devices III with a BPhen/Alq₃ multiple heterostructure ETL at an applied voltage of 11 V is above 14,000 cd/m², and the maximum luminance efficiency of device III at 273 mA/cm² is 4.5 cd/A. The BPhen/Alq₃ multiple heterostructure ETL in device III increases electron injection due to the high electron mobility of the BPhen layer. Because the electron mobility in the BPhen layer is larger than that in the Alq₃ layer, the space charges in the Alq₃ ETL dominantly affect the electron injection to the EML. Electron space charges existing in the Alq₃ ETL of device I at high voltages interrupt the electron injection to the EML, resulting in a decrease in the luminance and the luminance efficiency of device I. However, because the electron space charges in the multiple heterostructure ETL of device III are distributed into three regions due to the

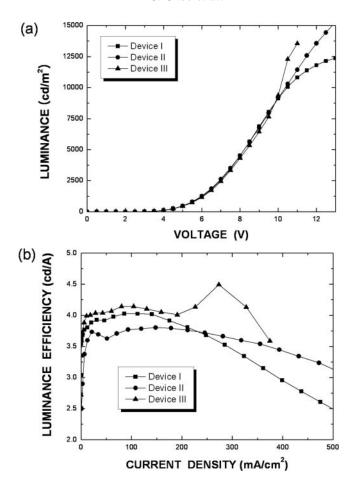


Figure 3. (a) Luminances as functions of the applied voltage, and (b) luminance efficiencies as functions of the current density for devices I, II, and III.

existence of the BPhen layer with a high electron mobility, the electrons injected from the ETL to the EML of device III are the largest value among all devices. Furthermore, the tunneling holes through the BPhen layer at high voltages recombines the accumulated electrons in the Alq₃ layer of the multiple heterostructure ETL, resulting in an increase in the luminance and the luminance efficiency of device III. The BPhen layer of device III efficiently blocks the hole transport from the EML to the ETL, resulting in the enhancement of the luminance efficiency in comparison with those of devices I and II. The luminance efficiency of device II is lower than that of device III because the BPhen layer cannot play an important role in blocking the hole transport from the EML to the ETL through the HOMO of the BPhen:Alq₃ mixed ETL.

Figure 4 shows EL spectra at 10 V for devices I, II, and III. The dominant peak of all devices appears approximately at 529 nm, and the full width at half maximum of the EL peak is approximately 97 nm. The dominant EL peaks of devices I, II and III are not shifted, regardless of an increase of the applied voltage. Because the formation and the recombination of excitons are only confined within the Alq₃ layer, any additional peak related to exciplex or excimer does not appear at EL spectra.

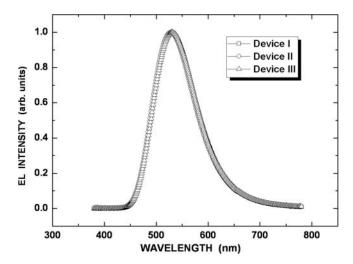


Figure 4. Electroluminescence spectra at 10 V for devices I, II, and III.

Conclusions

The electrical and the optical properties of OLEDs with three periods of BPhen/Alq₃ multiple heterostructures acting as an ETL were investigated. While the current density of OLEDs with a BPhen/Alq₃ multiple heterostructure ETL at low voltages was smaller than that of OLEDs with a BPhen:Alq₃ mixed ETL, the luminance efficiency of OLEDs with a BPhen/Alq₃ multiple heterostructure ETL was larger than that of OLEDs with a BPhen:Alq₃ mixed ETL. The maximum luminance efficiency of the OLEDs with a multiple heterostructure acting as an ETL at 273 mA/cm² was 4.5 cd/A. The enhancement of the luminance efficiency for the OLEDs with a BPhen/Alq₃ multiple heterostructure ETL originated from the high electron mobility and the hole blocking effect of the BPhen layer in the multiple heterostructure ETL. Because the formation and the recombination of excitons were only confined within the Alq₃ EML, an additional peak related to exciplex or excimer did not appear at the EL spectra. These results indicate that highly efficient OLEDs can be fabricated using a multiple heterostructure ETL consisting of the BPhen and the Alq₃ layers.

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