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Enhanced efficiency and reduced roll-off in nondoped phosphorescent organic light-emitting devices with triplet multiple quantum well structures

Shumei Liu,[1,2] Bin Li,[1,a)] Liming Zhang,[1] Hang Song,[1] and Hong Jiang[1]
[1]*Key Laboratory of Excited State Processes, Changchun Institute of Optics, Fine Mechanics, and Physics, Chinese Academy of Sciences, 3888-Dong Nan-Hu Road, Changchun 130033, People's Republic of China and Graduate School of the Chinese Academy of Sciences, Beijing 100039, People's Republic of China*
[2]*Department of Materials Science and Engineering, Jilin University, Changchun 130025, People's Republic of China*

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Highly efficient nondoped phosphorescent organic light-emitting devices (NPOLEDs) with triplet multiple quantum well structures are fabricated by using 4,4′-N,N′-dicarbazole-biphenyl and an iridium(III) complex as the potential barrier layer and the potential well layer/light-emitting layer, respectively. Remarkably, such NPOLED with an optimized device configuration achieves reduced current efficiency roll-off, which slightly decreases from its peak value of 31.5 cd/A at 19.8 mA/cm^2 to 29.2 cd/A at 100 mA/cm^2. We attribute this improvement to the efficient triplet exciton confinement effect and the suppression of triplet-triplet annihilation which occurs via single-step long range (Förster-type) energy transfer between excited molecules. © *2010 American Institute of Physics*. [doi:10.1063/1.3483131]

Since the pioneering works on phosphorescent organic light-emitting devices (PHOLEDs) reported by Forrest and co-workers,[1] PHOLEDs have attracted intensive attention due to the highly efficient emissions compared to conventional fluorescent counterparts. However, high-performance PHOLEDs are usually fabricated by using host-guest system to improve energy transfer and avoid triplet-triplet annihilation (TTA).[2–4] Realistically, this approach suffers from the poor reproducibility for mass production processes.[5–7] In addition, these devices exhibit significant efficiency roll-off at high current densities.[8,9] It is thus an important consideration on how to suppress the efficiency roll-off.

Multiple quantum well (MQW) structure has been proved to be an efficient way to improve device performances of organic light-emitting diodes (OLEDs). For example, Huang *et al.*[10] used a MQW structure to increase the carrier recombination efficiency, where both exciton and charge were confined to emitting layer (EML). Qiu *et al.*[11,12] improved hole-electron balance by utilizing an organic MQW structure to decelerate the hole transportation. Kim *et al.*[13] and Park *et al.*[14] also reported similar triplet MQW structures. In these reports, triplet MQW structures are fabricated by doping emitters into a charge-transporting host matrix. Recently, Divayana *et al.*[15,16] proposed a sequential doping method instead of the host-guest doping method. However, the device performances have no significant improvement compared with those of dopant OLEDs due to the negative effects caused by the too thin EML.

Here we report an efficient nondoped phosphorescent organic light-emitting device (NPOLED) by introducing a triplet MQW structure with *bis*(2-(2-fluorphenyl)-1,3-benzothiozolato-N,$C^{2'}$) iridium (acetylacetonate) [$(F\text{-}BT)_2Ir(acac)$] as the EML and potential well layer (PWL), while 4,4′-N,N′-dicarbazole-biphenyl (CBP) as the potential barrier layer (PBL). Charge carriers and triplet excitons are confined within each EML, which improves recombination efficiency and decreases current leakage. Furthermore, the CBP layer suppresses the single-step long range (Förster-type) energy transfer between excited molecules in adjacent well layers, reducing efficiency roll-off. The performances of our device are the best among reported $(F\text{-}BT)_2Ir(acac)$ dopant OLEDs.

The configuration of NPOLEDs with triplet MQW structure is indium tin oxide (ITO)/N,N'-bis-(1-naphthl)-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) (40 nm)/$[CBP(5\ nm)/EML(2\ nm)]_n$/4,7-diphenyl-1,10-phenanthroline (Bphen) (50 nm)/LiF (0.8 nm)/Al, and their schematic energy-level diagram is depicted in Fig. 1. Here, EML consists of $(F\text{-}BT)_2Ir(acac)$, *n* is well number ranging from 2 to 6. NPB is the hole transporting layer, while Bphen acts as the exciton blocking/electron transporting layer. The OLED with a structure of ITO/NPB (40 nm)/CBP: [$(F\text{-}BT)_2Ir(acac)$] (12 wt %, 30 nm)/Bphen (50 nm)/LiF (0.8 nm)/Al is introduced as a reference device (RD), hereafter. The emitting area is $2\times2\ mm^2$ in all devices. All organic layers and LiF/Al are deposited onto ITO-coated glass sub-

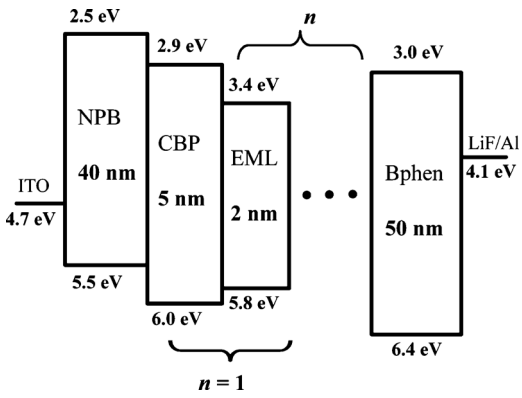

FIG. 1. Schematic energy-level diagram of NPOLEDs with triplet MQW structure. The HOMO level and the LUMO level were cited from literature Ref. 17.

a)Author to whom correspondence should be addressed. Electronic mail: lib020@yahoo.cn.

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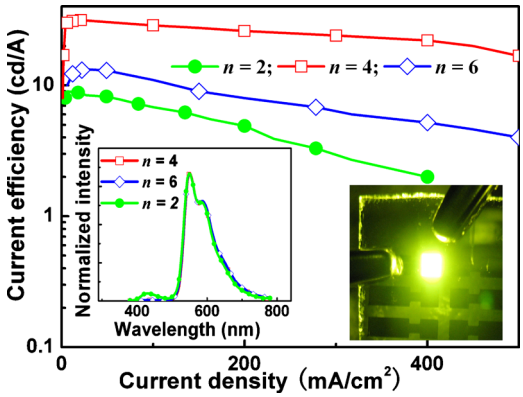

FIG. 2. (Color online) Current efficiency vs current density characteristics for NPOLEDs with different well number n. Left inset: EL spectra upon different well number n at 8 V. Right inset: A photograph of a pixel for NPOLED with $n=4$ during operation at 8 V.

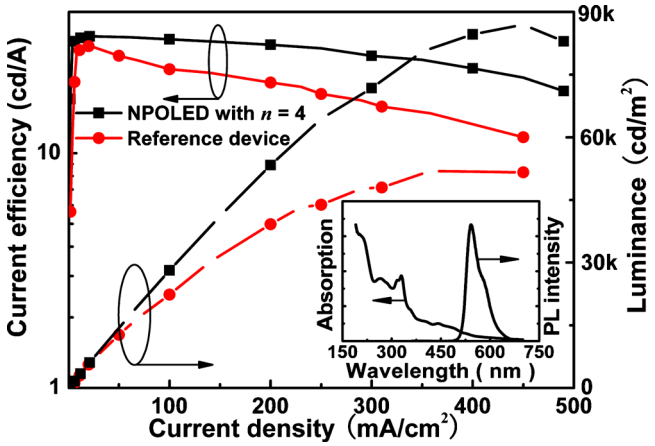

FIG. 3. (Color online) Current efficiency/luminance vs current density curves for MQW structure with $n=4$ and RD, respectively. Inset: Absorption and emission spectra of $(F\text{-}BT)_2Ir(acac)$ in deoxygenated CH_2Cl_2 solution (1×10^{-4} mol/l) at 298 K.

strates in vacuum under a chamber pressure of $\sim10^{-5}$ Pa. The electroluminescence (EL) spectra are measured with a PR650 spectrophotometer. The luminance-current versus voltage characteristics are measured simultaneously with a programmable Keithley 2400 voltage-current source. All measurements are carried out at room temperature under ambient conditions.

Figure 2 shows current efficiency versus current density curves for NPOLEDs upon various well numbers of n from 2 to 6. It is observed that the device with $n=4$ offers the highest current efficiency of 31.5 cd/A, with the minimum efficiency roll-off, which may be explained as follows. The highest occupied molecular orbital (HOMO) energy level of $(F\text{-}BT)_2Ir(acac)$ is 5.8 eV, 0.2 eV lower than that of CBP, meanwhile, the lowest unoccupied molecular orbital (LUMO) energy level of $(F\text{-}BT)_2Ir(acac)$ is 3.4 eV, 0.5 eV higher than that of CBP.[17] A deep electron trap and a shallow hole trap will thus be formed in each PWL/EML, efficiently capturing electrons and holes within PWL. Besides, the triplet energy of $(F\text{-}BT)_2Ir(acac)$ ($T_1=2.25$ eV) is lower than the corresponding ones of CBP ($T_1=2.56$ eV) and BPhen ($T_1=2.59$ eV), preventing triplet excitons leakage out of EML.[17,18] Therefore, charges and triplet excitons can be effectively confined in the EML, resulting in a high recombination efficiency. The hole mobility of CBP and NPB are 2×10^{-3}, and 1×10^{-3} $cm^2\ V^{-1}\ s^{-1}$, respectively, which are comparable to the electron mobility of CBP (2×10^{-4} $cm^2\ V^{-1}\ s^{-1}$) and BPhen (1×10^{-4} $cm^2\ V^{-1}\ s^{-1}$).[19,20] It is thus expected that charge transportation balance can be achieved within MQW layers. Furthermore, the thin EML blocks hole transporting to PBL due to the electron-favoring character of $(F\text{-}BT)_2Ir(acac)$. The 0.2 eV energy barriers at EML/CBP interface may also limit the hole transporting process. In consequence, the fast-moving holes may slow down in EML, further improving the charge transportation balance of holes and electrons. When $n=2$, electrons can easily either cross or tunnel through the 0.5 eV PBLs to reach the NPB/CBP interface, leading to a charge recombination on NPB, which is confirmed by the emission band peaking at 450 nm shown in the left inset of Fig. 2. The main band peaking at 548 nm is comparable with the reported one of 550 nm in dopant OLED,[21] indicating that aggregation effect in NPOLEDs is negligible. When $n=4$ and 6, no NPB emission is detected, meaning that no electrons leak out of EML. We attribute the rather low device efficiency of $n=6$ to the charge transportation imbalance and the excess interfaces which may cause negative effects such as defect state or nonradiative center.[10,22] A photograph of bright yellow-green emission originated from a pixel of NPOLED with $n=4$ at 8 V is shown in the right inset of Fig. 2.

Figure 3 shows the efficiency/luminance versus current density characteristics of the NPOLED with $n=4$ and RD. The device efficiency values of RD are found to be 28.5 cd/A at 20 mA/cm^2, 22.8 cd/A at 100 mA/cm^2, and 15.6 cd/A at 400 mA/cm^2, respectively. While, the NPOLED with $n=4$ surpasses RD by showing corresponding device efficiency values of 31.5 cd/A at 19.8 mA/cm^2, 29.2 cd/A at 100 mA/cm^2, and 22.5 cd/A at 400 mA/cm^2. In addition, the device efficiency decreases more smoothly with the increase in current density compared with that of RD. The NPOLED with $n=4$ exhibits a maximum luminance of 85 700 cd/m^2 upon a current density of 450 mA/cm^2, 1.6 times higher than that of RD (52 000 cd/m^2). We give the causation of such excellent performances as follows. For most PHOLEDs, owing to hole's easy injection from anode and the higher mobility, excitons are formed at the interface of EML and hole-blocking layer. As current density increases, the recombination zone may shift and become wider, resulting in a sharp decrease in device efficiency.[18] Confining charge and exciton within EML has been reported to be an effective method to reduce the rapid dropping of device efficiency at high current densities.[13,14] In our MQW devices, charges and excitons are effectively confined to EML due to the wide band gap and the high triplet energy of CBP PBL. In contrast, NPB emission can be observed at high current densities in RD for electron diffusing into the interface of NPB/CBP layers.[21] Besides, the overlap between emitting materials absorption and emission bands may trigger single-step long range Förster-type transfer, resulting in intense triplet–triplet annihilation.[15,23] The Förster radius in most PHOLEDs is found to be around 4 nm.[15] As shown in the inset of Fig. 3, the emission spectrum of complex $(F\text{-}BT)_2Ir(acac)$ centering at 542 nm overlaps significantly with its triplet metal to ligand charge transfer (^{3}MLCT) absorption ranging from 500 to 650 nm, indicating a potential intermolecular energy transfer process between excited $(F\text{-}BT)_2Ir(acac)$ molecules.[21] Correspondingly, in RD, adja-

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cent emitters are so close to each other that TTA originating from Förster-type self-quenching may happen, leading to the sharp efficiency roll-off. As for NPOLEDs with MQW structures, however, the emitting zone is periodically interrupted by 5 nm of PBL, effectively preventing such TTA, and then the efficiency roll-off was reduced.

In summary, a high efficiency NPOLED with reduced efficiency roll-off is fabricated by using a nondoped triplet MQW structure. The current efficiency slightly changes from its peak value of 31.5 cd/A at 19.8 mA/cm^2 to 29.2 cd/A at 100 mA/cm^2, remaining over 90% of its peak efficiency. We attribute such high efficiency and reduced efficiency roll-off at high current densities to the following three advantages caused by the triplet MQW structure: (1) efficient charge and exciton confinement effect by PBL; (2) charge transportation balance in each PWL/EML; (3) suppression on the long-range Förster-type self-quenching process. The described MQW device concept may be useful to fabricate highly efficient NPOLEDs with low efficiency roll-off for future OLED display and lighting applications.

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