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High-purity white light from a simple single dopant host-guest white organic light-emitting diode architecture

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White light with good color properties (color rendering index=82–87) is generated in a simple organic light-emitting diode comprising an emissive layer, composed of an undoped *tris*(4-methyl-8-quinolinato)aluminum ($Almq_3$) sublayer and region doped with an orange-red phosphorescent dopant, bis(2-phenyl-1-quinoline)iridium acetylacetonate ($Ir(pq)_2acac$). Electron-hole recombination in a thin spacing $Almq_3$ layer results in blue-green fluorescence, while the formed triplet excitons diffuse to the doped region and are harvested by the dopant to emit orange-red phosphorescence. The combination of blue-green and orange lights results in warm white light. This approach takes advantage of efficient migration of triplet excitons while being less demanding in terms of fabrication and color matching. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3005424]

The development of efficient white organic light-emitting diodes (WOLEDs) for general lighting purposes has been intensively pursued over the last few years. In order to become a competitive technology in solid-state lighting, a WOLED light source should be more efficient than incandescent (approximately 16 lm/W; 100 W) and fluorescent tube (approximately 60 lm/W; 32 W) light sources, display high color rendering index (CRI>80), and should be stable over long operational lifetime.[1] Several approaches have been attempted to realize this goal including the fabrication of stacked architectures,[2] devices with separate emitter layers,[3] use of excimer and/or exciplex emission,[4] as well as broadband emitting copolymers.[5]

Recent results suggest that WOLED prototypes can indeed achieve high efficiencies while generating high quality white light.[1] The highest external quantum efficiency (EQE) obtained for a device with excimer-based electroluminescence (EL) was reported by Jabbour *et al.*: 15.9% at a luminance of 500 cd/m^2.[4(b)] However, the excimer approach usually affords very poor color rendering which is also the case for this example (CRI<75). The incorporation of three different emitters into the emissive layer has allowed making efficient use of singlet and triplet excitons and impressive results have been obtained. An EQE of 10.8% with a CRI of 85 was achieved by Sun *et al.* at a luminance of 500 cd/m^2,[6] while Schwartz *et al.* have recently reported efficiencies of 12.9% and 14.1% (CRI=86 and 82, respectively) at 1000 cd/m^2 by incorporating microlens outcoupling foil.[7] Unfortunately, the above approaches require careful adjustment of each emissive layer in order to balance the relative intensity of the EL, thus increasing the attendant complexity of the fabrication process and the cost of the device.

Producing high-CRI (>85%) white light using simple WOLED configurations would be of significant advantage for high volume fabrication associated with potential general lighting applications. Toward this end, we decided to further explore the possibility of generating white light by combining blue-green emission from a fluorescent host with orange-red phosphorescence. The materials *tris*(4-methyl-8-quinolinato)aluminum ($Almq_3$) and bis(2-phenyl-1-quinoline)iridium acetylacetonate ($Ir(pq)_2acac$) were selected to display broad emission to cover as much of the visible light spectrum as possible to generate white light.[8] This device is a simple analogy of the devices by Sun *et al.*[6] and Schwartz *et al.*[7] in the sense that it also utilizes exciton diffusion to generate electrophosphorescence from a doped layer spatially separated from the recombination zone. In the present devices, the emissive layer is composed of two emissive sublayers: an undoped $Almq_3$ host sublayer and a $Almq_3$ host region doped with an orange dopant (5% $Ir(pq)_2acac:Almq_3$). Due to the low hole mobility in the host, the charge recombination is likely to take place close to the 4,4'-bis[*N*-(1-naphtyl)-*N*-phenylamino]biphenyl (NPD)-$Almq_3$ interface resulting in singlet excitons and the blue-green light emitted from the $Almq_3$. The triplet excitons generated diffuse through this undoped host spacer layer (5.0–15.0 nm) and are harvested[6,7] by the second sublayer composed of the $Ir(pq)_2acac$ dopant in $Almq_3$ (30 nm). The spectral overlap of the $Almq_3$ emission and $Ir(pq)_2acac$ metal-to-ligand charge transfer absorption enable, in principle, the fluorescence resonance energy transfer (FRET) from $Almq_3$ to the $Ir(pq)_2acac$ dopant. If the $Almq_3$ sublayer is too thin, the singlet excitons formed at the NPD-$Almq_3$ interface are within the Förster radius (3–6 nm) (Ref. 9) distance for FRET. In such a case, the singlet excitons would be harvested by the dopant and followed by the intersystem crossing and red phosphorescence. As a result, the electroluminescence profile would lose the blue-green component. For this reason, we evaluated several thicknesses of the $Almq_3$ layer adjacent to the hole-transporting layer (HTL). Low thickness of the $Almq_3$ spacing layer gives poor CRI and a comparable EQE, while high thickness of the $Almq_3$ layer results in a light output with a blue hue, poor CRI, and lower EQE, the latter presumably due to the loss of triplet excitons due to the incomplete diffusion harvesting over a relatively long distance (~15.0 nm). The thickness of the $Almq_3$ spacing layer x was varied between 5.0 and 15.0 nm. Thickness x of approximately 10 nm was found to be a good

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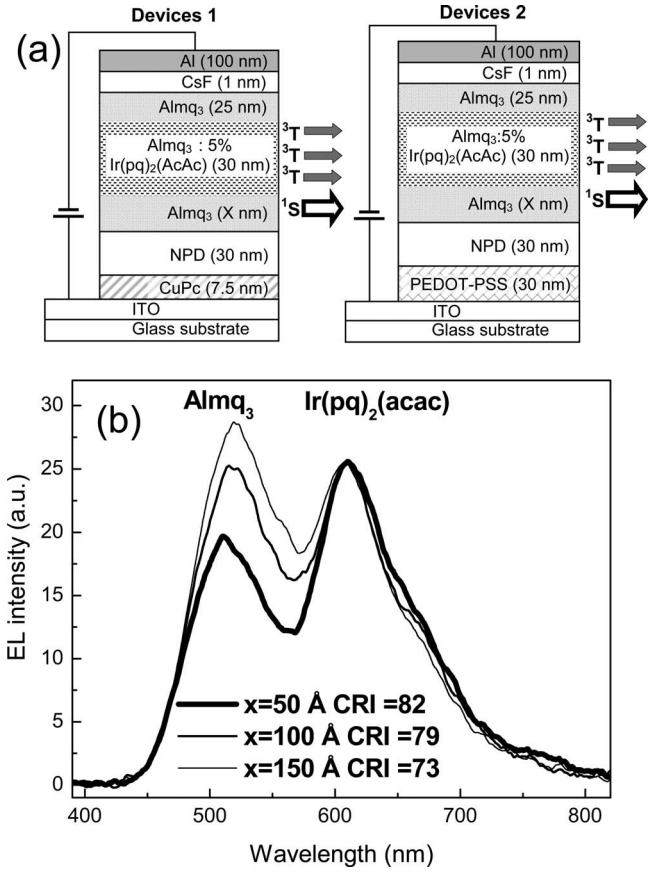

FIG. 1. (a) Structure of the devices with varying thickness of the $Almq_3$ fluorescent emissive layer. (b) EL spectra for devices of type 1 with X =5.0–15.0 nm at 80 mA/cm^2.

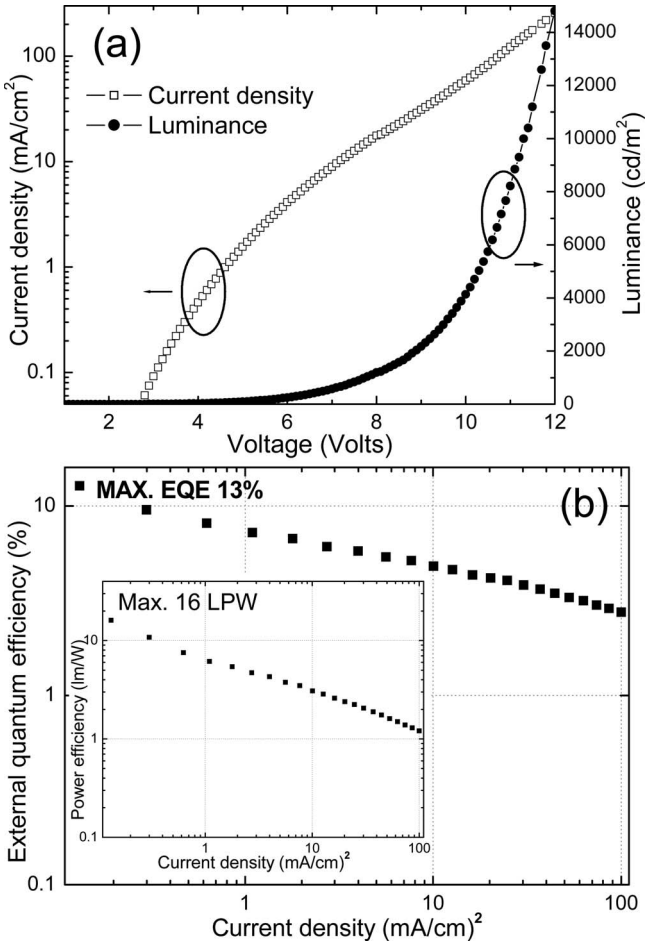

FIG. 2. (a) Luminance and J vs V curves for device 2. (b) Dependence of the EQE on the current density of the diode. The inset shows power efficiency vs current density.

compromise. The spacer layer of the $Almq_3$ host thus produces the blue-green fluorescence, prevents dipole-dipole energy transfer, and diminishes triplet-polaron quenching effects by having the dopant dispersed into the electron-transporting layer.[10] Two types of devices were fabricated: devices 1, which utilize CuPc as a hole-injection layer, and devices 2 with poly(3,4-ethylenedioxythiophene)/poly-(strene sulfonic acid) (PEDOT-PSS) (Clevios P and PH 500) to planarize the indium tin oxide surface and to prevent pinhole defects.

For the devices 1 and 2, CuPc and PEDOT:PSS, respectively, were used as a hole-injection layer (30 nm), followed by a 4,4′-bis[N-(1-naphtyl)-N-phenylamino]biphenyl (α-NPD;30 nm) hole-transporting layer. The emissive layer composed of two sublayers [$Almq_3$ and 5% $Ir(pq)_2acac:Almq_3$] was followed by an $Almq_3$ electron transport layer and cesium fluoride (CsF; 1 nm) as the electron injection layer and aluminum (Al) cathode. For all the devices, the EL spectra showed contributions from both emitters with peaks at 510 and 610 nm as expected. The thickness of the $Almq_3$ spacing layer determines to a large degree the color output of the device. For example, in device 1 (CuPc), for the thicknesses of $Almq_3$ (X) of 5, 10, and 15 nm, the corresponding CRI values recorded were 82, 79, and 73, as a result of increased contribution of the $Almq_3$ emission in the EL spectra (see Fig. 1).

In order to aid hole injection and avoid the potential of electrical shorts,[11] we introduced a 300 Å PEDOT:PSS (Clevios) layer (device 2). The PEDOT:PSS-α-NPD devices 2 were expected to yield a slightly better overall performance. For device 2 with $Almq_3$ spacer layer X=10.0 nm, we recorded maximum EQE of 13%. At the luminance of 500 cd/m^2, which is practical for lighting purposes the recorded efficiency was 6%–7% and the current density was ~4 mA/cm^2. Interestingly, device 2 displayed a slightly higher contribution from $Ir(pq)_2acac$ in the EL spectra and a high general CRI of 87, Commission Internationale de I'Eclairage (CIE) coordinates X, Y of 0.457 and 0.450, and a color temperature of 3030 K at 500 cd/m^2. The turn-on voltage is only 3 V and the voltage required to reach 1000 cd/m^2 is less than 7.5 V. Figure 2 displays the current-voltage-luminance curves as well as the EQE and the power efficiency as a function of the current density. These data describe a simple yet very efficient WOLED, with high color rendering, which is remarkable considering the simplicity of this device.

A proof that a diffusion-based harvesting of the triplet excitons is taking place is provided by the fact that an undoped $Almq_3$ device displayed an EQE of only ~1.8% at 500 cd/m^2,[12] which implies that the improvement in the efficiency for the doped OLED must originate from efficient utilization of the triplet excitons generated at the HTL-host interface. The white emission obtained from both types of devices showed very weak color dependence for luminances between 150 and 2000 cd/m^2, a range practical for lighting purposes (see Fig. 3, left). This result demonstrates the ad-

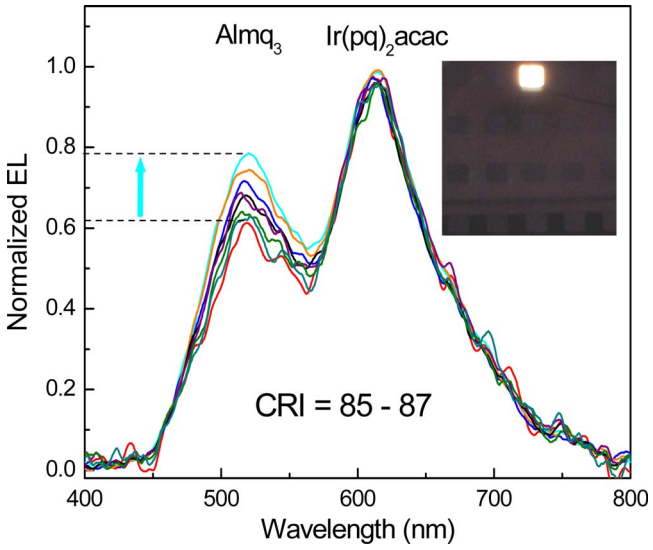

FIG. 3. (Color online) EL spectra recorded at various luminances between 150 and 2000 cd/m^2 showing small variation in the white light characteristics of device 2. Inset: Photograph of one of the pixels operating at 6 V.

vantage of this approach where only one dopant is employed to complement the emission of a fluorescent host layer. We believe that this approach can be extended to other electron-transporting layer (ETL) materials and emitters such as blue-emitting $Almq_3$ derivatives[13] and other orange-red phosphorescent emitters [i.e., $Os(fptz)_2(PPh_2Me)_2$].[14]

In conclusion, we demonstrate that through a simple approach it is possible to efficiently generate white light utilizing simple architecture where the emissive layer comprises an undoped *tris*(4-methyl-8-quinolinato)aluminum ($Almq_3$) sublayer and a region doped with an orange-red phosphorescent dopant, bis(2-phenyl-1-quinoline)iridium acetylacetonate ($Ir(pq)_2acac$). The electron-hole recombination in a thin spacing $Almq_3$ layer results in blue-green fluorescence, while the formed triplet excitons diffuse to the doped region where they are harvested by the dopant to emit orange-red phosphorescence, which together furnish white light.

This simple method is significantly less demanding in terms of color matching of multiple doped layers while providing excellent-color rendering (CRI=87), high luminous output, and reasonable efficiency. By spatially separating the exciton generation zone from the doped layer, we were able to achieve balanced white EL with a maximum EQE of 13% (EQE of 10.0% in at a current density of 0.3 mA/cm^2). At the luminance of 500 cd/m^2, which is practical for lighting purposes the recorded efficiency was 6%–7% and the current density was ~4 mA/cm^2. We believe that using this scheme the choice of different host-dopant combinations might result in higher efficiencies and that a better performance could be obtained by improving charge injection (i.e., doping with $TF_4TCNDQM$ to reduce operating voltage and increase the power efficiency).[15] Studies toward this end are the subject of ongoing research.

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