Efficient White OLEDs Employing Phosphorescent Sensitization

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Abstract—We have investigated white-emitting organic light-emitting devices (WOLEDs) making use of both blue-phosphor-sensitized orange-red fluorescence and the residual blue phosphorescence. By carefully adjusting the concentrations the phosphor and the fluorophore in the emitting layer and choosing the carrier-transport layers in the device structure, WOLEDs containing a single phosphor-sensitized emitting layer (type-I devices) can give colors close to the equal-energy white (0.33, 0.33), CRI up to 75, and efficiencies up to (10%, 23 cd/A, 13.4 lm/W). Furthermore, by doping a green phosphor into the poorly emitting electron-transport layer (type-II devices) to recycle excitons formed there, the EL efficiencies can be further enhanced up to (12.1%, 35.3 cd/A, 23.9 lm/W). In both types of devices, the phosphor sensitization reduces population of triplet excitons in the emitting region and substantially mitigates the efficiency roll-off with the driving current or brightness that is often observed in all-phosphor OLEDs. At the brightness of 1000 cd/m², both types of devices retain quantum and cadmium per ampere (cd/A) efficiencies similar to their peak values.

Index Terms—Phosphorescent sensitization, solid-state lighting, white organic light-emitting devices (WOLEDs).

I. INTRODUCTION

DUE to continuous improvement of people's living, usage of energy continues to increase, while various energy security measures indicate the potential of an energy shortage [1], [2]. On one hand, scientists worldwide are seeking new replacement resources to alleviate such an issue. On the other hand, improving the efficiency of energy usage is more economical and environment-friendly, and thus should be put into practice with high priority. Generally speaking, among various uses of energy, electricity for lighting accounts for nearly 10% of the total energy consumption [1], [2]. Therefore, the development and use of high-efficiency solid-state lighting to replace conventional lighting sources is one of the most effective energy-saving strategies. Organic light-emitting devices (OLEDs), due to their potentially high power efficiencies, their surface-emitting characteristics (thus no need for substantial assembly), their mechanical flexibility, and their capability to be fabricated on the

conformable and flexible substrate for many new applications, are considered as one of the most promising next-generation lighting technologies.

Since Kido's group reported the first white organic light-emitting devices (WOLEDs), more and more researchers are getting involved in the development of high-efficiency white organic electroluminescence (EL) [3]–[10]. In recent years, with the development and use of efficient organic phosphorescent emitters, efficiencies of WOLEDs are subjected to great enhancement [8]–[10]. Organic phosphorescent emitters containing transition metals renders possible harvesting both electro-generated singlet and triplet excitons for emission and realizing nearly 100% internal quantum efficiencies of electroluminescence [11].

Typically, emission of typical organic materials only spans about one third of the visible spectrum, and thus the white light must be obtained by mixing two complementary colors or three primary colors to span a broad white emission. Intuitively, high-efficiency WOLEDs could be accomplished by employing all-phosphor doped systems [8]–[10]. In phosphorescent OLEDs, in general high concentrations of phosphorescent dopants in host layers are needed for achieving efficient short-range Dexter energy transfer and for obtaining high efficiencies. As such, phosphorescent OLEDs usually suffer rapid roll-off of efficiencies at high excitation densities (i.e., high concentrations of triplet excitons), which is associated with long lifetimes of triplet excitons and the triplet-triplet annihilation [8]–[13]. Such an issue will be particularly critical for WOLEDs since for lighting applications WOLEDs will be typically operated at high driving currents and high brightnesses. This issue may be mitigated by phosphor-sensitized fluorescence demonstrated by Baldo *et al.* [12], [13], in which resonant energy transfer occurs between triplet excitons in the phosphor and singlets in the fluorophore. In the phosphor-sensitized system, a conductive host is doped with both phosphors and fluorophores. With doping a phosphor at high concentrations into a conductive host, both singlet and triplet excitons can transfer onto the phosphor molecule, which are then all transferred to the radiative triplet excited states of the phosphor if strong spin-orbit coupling exist to facilitate intersystem crossing. The radiative triplet states of the phosphor can then be readily transferred via the long-range dipole-dipole Förster process to the radiative singlet state of the fluorophore [12], [13]. With low doping of the fluorophore, the undesired transfer from host/phosphor triplets to the nonradiative triplet state of the fluorophore (through the Dexter process) is discouraged. Thus in principle, phosphor sensitization can lead to 100% internal quantum efficiency of OLEDs from fluorescence.

In Baldo's studies [12], [13], the efficient green phosphor Ir(ppy)₃ is used as the sensitizer for yellow or orange-red fluo-

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Fig. 1. Molecular structures of organic materials used.

Fig. 2. Schematic structures of type-I and type-II devices.

rescent dyes such as DCM2. The resulting emission is yellow/orange-red emission from the fluorescent dye. Later in 2004, Lei *et al.* demonstrated that the white-emitting OLEDs could be obtained by using a blue phosphor FIrpic as the sensitizer and the orange-red dye DCJTb as the fluorophore [14]. By carefully adjusting the relative concentrations of the phosphor and the fluorophore, the emission from the device combined both residual blue phosphorescence from the sensitizer FIrpic and the sensitized orange-red fluorescence from DCJTb, giving white EL. This approach for generating white EL is attractive, yet Lei *et al.* achieved a peak current efficiency of only 9.2 cd/A, which is substantially lower than those of state-of-the-art all-phosphor WOLEDs [8]–[10].

Since the blue phosphorescent emitter FIrpic in a wide-gap host in principle can exhibit a very high photoluminescence (PL) quantum efficiency (> 90%) [15], there should be still plenty of room in improving EL efficiencies of WOLEDs making use of both phosphorescence of FIrpic and FIrpic-sensitized fluorescence. In this work, we investigate the device structures of such

WOLEDs and show that a substantially enhanced efficiency of over 30 cd/A indeed can be achieved.

II. EXPERIMENTAL

A. Device Structures

Using the materials shown in Fig. 1, two types of devices (Fig. 2) were fabricated and tested. In type-I devices, a single phosphor-sensitized emitting layer was used, while in type-II devices, two emitting layers, one phosphor-sensitized emitting layer and one phosphorescent emitting layer were used.

The configuration of the type-I devices is: ITO/PEDOT:PSS (~ 30 nm)/TCTA (30 nm)/mCP:FIrpic (8 wt.%):DCJTb (*x* wt.%) (30 nm)/TAZ (40 nm)/LiF (0.5 nm)/Al (150 nm). The emitting layer (EML) consists of the 1,3-bis(9-carbazolyl)benzene (mCP) [16] host co-doped with the blue phosphorescent complex bis[(4,6-difluorophenyl)-pyridinato-*N*, *C*²](picolinate)Ir(III) (FIrpic) and the orange-red fluorescent dye 4-(dicyanomethylene)-2-*t*-butyl-6-(1,1,7,7-tetramethyl-2,3,8,9-tetrahydronaphtho[2,3-*b*]pyridin-2-yl)-2,3,8,9-tetrahydronaphtho[2,3-*b*]pyridine (DCJTb).

ethyljulolidyl-9-enyl) (DCJTB) of various concentrations (0–0.5 wt.%). The low concentration of DCJTB is adjusted to retain the partial blue phosphorescence of Flrpic and yet also obtain sensitized orange-red fluorescence of DCJTB. The conducting polymer poly(3,4-ethylenedioxythiophene)/poly(styrene sulfonic acid) (PEDT:PSS) is spun onto the indium-tin-oxide (ITO)-coated glass substrate to serve as the hole injection layer [17]–[19]. 4,4',4''-tris(carbazole-9-yl)-triphenylamine (TCTA) and 3-(4-Biphenyl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole (TAZ) are used as hole-transport layer and the electron-transport layer (HTL and ETL), respectively [17]–[19]. The cathode consists of a LiF electron-injection layer and Al.

The configuration of the type-II devices is: ITO/PEDT:PSS (~ 30 nm)/TCTA (30 nm)/mCP (10 nm)/mCP:Flrpic (8 wt.%):DCJTB (0.15 wt.%) (20 nm)/TAZ:Ir(ppy)₃ (y wt.%, 10 nm)/TAZ (30 nm)/LiF(0.5 nm)/Al(150 nm). This device structure is similar to that of type-I devices except that the DCJTB concentration is fixed at 0.15 wt.% and an additional green phosphorescent emitter *fac*-tris(2-phenylpyridine)Ir(III) (Ir(ppy)₃) [8]–[10] with high quantum efficiency and varied concentrations is doped into the region of the electron-transport layer (TAZ) adjacent to the phosphor-sensitized emitting layer. Thus type-II devices contain two emitting layers, whose total thickness is still fixed at 30 nm as in the type-I devices.

B. Device Fabrication and Testing

Prior to deposition of the organic layers, the ITO-coated glass substrates were cleaned with the detergent, deionized water and organic solvents, and then treated with UV ozone [17]–[19]. The conducting polymer was deposited by spin-coating. Other material layers (organic and inorganic) were deposited by vacuum evaporation in a vacuum chamber with a base pressure of $< 10^{-6}$ torr. The deposition system permits the fabrication of the complete device structure in a single pump-down without breaking vacuum.

Current-voltage-brightness (I - V - L) characterization of the light-emitting devices was performed with a source-measurement unit (SMU) and a Si photodiode calibrated with Photo Research PR650. EL spectra of devices were measured by a calibrated spectrometer with a charge-coupled device (CCD) array detector.

III. RESULTS AND DISCUSSION

A. Type-I Devices With a Single Phosphor-Sensitized Emitting Layer

Fig. 3 shows the EL spectra of type-I devices, in which all the spectra are normalized with respect to the emission peak of Flrpic. The concentration of Flrpic in mCP was fixed at 8 wt.% and the concentration of DCJTB was varied from 0 to 0.5 wt.%. The EL spectrum of the control device without DCJTB shows mainly blue emission of Flrpic (with peaks at 470 and 495 nm). By increasing the concentration of DCJTB, the orange-red emission from DCJTB grows in relation to Flrpic emission. Such a phenomenon results from the enhanced resonant energy transfer from Flrpic to DCJTB since the triplet-to-singlet Förster transfer rate is proportional to the concentration of

Fig. 3. EL spectra of type-I devices with various DCJTB concentrations (at 100 mA/cm²).

Fig. 4. CIE coordinates of type-I devices with various DCJTB concentrations.

the acceptor [12], [13]. A bathochromic shift of the DCJTB peak from 580 to 600 nm is also observed with increasing the DCJTB concentration, which may be due to the solid-state solvation effect associated with the high polarity of DCJTB molecules [20], [21]. The 1931 CIE coordinates of OLEDs with various DCJTB concentrations, calculated from EL spectra, are summarized in Table I and are also shown in Fig. 4. The CIE coordinates of these OLEDs shift from (0.17, 0.31) of Flrpic emission to reddish white of (0.48, 0.41) with increasing the DCJTB concentration. A white color of (0.28, 0.36)–(0.35, 0.38), closest to the equal-energy white (0.33, 0.33), is obtained with a DCJTB concentration of 0.1–0.2 wt.%. The color shift of these WOLEDs with the bias voltage is considered small. For instance, for the device with 0.1 wt.% DCJTB, only a slight shift of the CIE1931 coordinates from (0.29, 0.37) to (0.28, 0.36) is observed when the voltage increases from 7 V (100 cd/m²) to 12 V (10000 cd/m²).

In addition to the CIE coordinates, there are two more parameters, i.e., the color-rendering index (CRI) and the correlated color temperature (CCT) [22], that are related to the color quality of WOLEDs for lighting applications. An ideal white-light source for lighting should have a high color rendering ability (i.e., with a CRI close to 100). In addition, colors of high-

TABLE I
DEVICE PERFORMANCES OF TYPE-I DEVICES

DCJTb (wt.%)		0	0.1	0.2	0.3	0.4	0.5
External Quantum Efficiency (%)	Maximum	10.7	10.0	8.0	6.2	4.8	4.5
	@ 100 cd/m ²	10.0	9.9	8.0	5.7	4.2	3.6
Luminance Efficiency (cd/A)	Maximum	22.8	22.9	19.8	13.8	13.7	9.6
	@ 100 cd/m ²	21.3	22.4	19.1	12.7	12.1	7.6
Power Efficiency (lm/W)	Maximum	10.8	13.4	12.4	8.6	8.4	6.0
	@ 100 cd/m ²	8.0	10.4	8.6	4.8	3.9	2.5
CIE 1931 Coordinates (x, y)		(0.17,0.31)	(0.28,0.36)	(0.35,0.38)	(0.43,0.41)	(0.44,0.40)	(0.48,0.41)
Color Rendering Index		—	62	72	75	73	71
Correlated Color Temperature (K)		—	8400	5200	3700	3500	2700

Fig. 5. External EL quantum efficiencies of type-I devices with various DCJTb concentrations.

quality lighting sources should be similar to those of Planckian radiators having CCT of 2500–6500 K. CRI and CCT of all type-I devices, calculated from EL spectra, are summarized in Table I. The CRI and CCT of the most efficient WOLED (with 0.1 wt.% DCJTb) are 62 and 8400 K, respectively, where the weaker orange-red component in EL renders the device emission bluish white and results in a lower CRI. As the concentration of DCJTb increases, the device emission becomes more warmish white. Higher CRI of 75 can be obtained with a DCJTb concentration of 0.3 wt.%.

The characteristics of external EL quantum efficiencies versus driving current for all type-I devices are shown in Fig. 5. Also in Table I, the external EL quantum efficiencies (maximal and at 100 cd/m²), cd/A efficiencies (maximal and at 100 cd/m²), and power efficiencies (maximal and at 100 cd/m²) of all type-I devices are summarized. The control blue phosphorescent device without DCJTb has a peak external EL quantum efficiency of 10.7% (22.8 cd/A) (Table I, Fig. 5). For the phosphor-sensitized type-I WOLEDs, the device with 0.1 wt.% DCJTb exhibits the highest peak external quantum efficiency of 10% (23 cd/A). The current-voltage-luminescence (*I-V-L*) characteristics of this device are shown in Fig. 6(a), while its external quantum efficiency and the power efficiency vs. current are shown in Fig. 6(b). This device has peak efficiencies of (13.4 lm/W, 10.0%, 23 cd/A) at a lower luminance. At the more practical brightnesses of 100 cd/m² and 1000 cd/m², this device exhibits efficiencies of (10.4 lm/W, 9.9%, 22.4 cd/A)

Fig. 6. (a) *I-V-L* characteristics. (b) External quantum efficiency/power efficiency versus current density for the type-I device with 0.1 wt.% DCJTb.

and (7 lm/W, 9%, 20.7 cd/A), respectively. It is worth mentioning that the EL efficiencies up to 23 cd/A and 10% here are substantially higher than that (9.2 cd/A) of Lei's devices [14], which were also based on FIrpic and DCJTb. Such an efficiency enhancement is mainly due to the improvement EL efficiency of the blue phosphorescent itself (22.8 cd/A in our FIrpic devices versus 9.8 cd/A in Lei's pure FIrpic device). On one hand, the host material used in our device is different from that used in Lei's device. On the other hand, hole-transport and electron-transport materials in our devices (TCTA and TAZ) are also different from those used in Lei's devices (N,N'-diphenyl-N,N'-bis(1,1'-biphenyl)-4,4'-diamine (NPB) and 4,7-diphenyl-1,10-phenanthroline (BPhen)). In our previous work on blue phosphorescent OLEDs, we have noticed that for the wide-gap host materials, using TCTA and TAZ as the charge-transport materials will give substantially better

device performances than using NPB and phenanthroline-based materials [17].

It is also interesting to note that the type-I device with 0.1 wt.% of DCJTB exhibits an EL quantum efficiency very similar to that of the control FIrpic device. This indicates that with a low DCJTB concentration (e.g., 0.1 wt.%), the undesired energy transfer from FIrpic triplets to the nonradiative triplet state of DCJTB (through the Dexter process) is discouraged, and the triplet energy of FIrpic is mainly transferred via the long-range dipole-dipole Förster process to the radiative singlet state of DCJTB. It is also worthy of noting that unlike purely phosphorescent devices, the current phosphor-sensitized device exhibits a much mitigated EL efficiency roll-off with the current and brightness (e.g., comparing efficiencies of the pure FIrpic device and the sensitized device in Fig. 5). For the 0.1-wt.%-DCJTB device, the EL efficiencies at 100 cd/m² and 1000 cd/m² are very similar to the peak EL efficiencies at lower brightnesses. This may be associated with the reduced population of triplet excitons on FIrpic (and thus reduced probability of triplet-triplet annihilation) since a portion of triplet excitons is transferred to singlets of DCJTB (which have much shorter excited-state lifetimes), seemingly an advantage of the phosphor-sensitized OLEDs compared to all-phosphor devices [12], [13].

As shown in both Fig. 5 and Table I, by increasing the DCJTB concentration in type-I devices from 0.1 wt.% to 0.5 wt.%, the EL efficiency gradually drops from 10% to 4.5%. The increase of the DCJTB concentration could result in rapid enhancement of the undesired energy transfer from FIrpic triplets to the non-radiative triplet state of DCJTB (through the Dexter process), since the Dexter process has an exponential dependence on the inverse of the donor-to-acceptor distance. This would lead to degradation of the overall EL efficiency. Furthermore, the concentration quenching of the DCJTB dyes may also partly contribute to loss of EL efficiency at higher DCJTB concentrations [20], [21].

B. Type-II Devices With Two Emitting Layers

In EL spectra of type-I devices (Fig. 3), in addition to emission from FIrpic and DCJTB, weaker emission ranging from 350 to 410 nm is also noticed. By inspecting the photoluminescence spectra of TCTA, TAZ and mCP, this emission can be unambiguously assigned to fluorescence of the electron-transport layer TAZ. The observation of TAZ emission suggests a portion of excitons may be formed on the TAZ side of the mCP/TAZ interface, perhaps due to some holes crossing this interface and some electrons being blocked by this interface. Since TAZ is not an efficient emitter (in both fluorescence and phosphorescence), formation of excitons on TAZ would lead to loss of device EL efficiency. Since the triplet energy of TAZ (~ 2.55 eV in thin films) is larger than typical green phosphorescent emitters [11], [17], it is possible to recycle the excitons on TAZ for more efficient emission by doping a green phosphorescent emitter in the region of the TAZ layer adjacent to the mCP/TAZ interface. Therefore, with slight modification of the device structure of type-I devices, type-II devices with doping the green phosphor Ir(ppy)₃ into TAZ near the interface (10 nm) were fabricated and tested.

Fig. 7. EL spectra of type-II devices with various FIrpic concentrations in TAZ (at 100 mA/cm²).

Fig. 8. CIE coordinates of type-II devices with various FIrpic concentrations in TAZ.

In type-II devices, the concentrations of FIrpic and DCJTB in mCP were fixed at 8 and 0.15 wt.%, respectively, while the Ir(ppy)₃ concentration in TAZ was varied from 0.3 wt.% to 1.0 wt.%. Fig. 7 shows the EL spectra of type-II devices with various Ir(ppy)₃ concentrations, in which all the spectra are normalized with respect to the emission peak of FIrpic. The corresponding 1931 CIE coordinates of these devices are shown in Fig. 8 and in Table II. From the EL spectra, it is clearly seen that by doping Ir(ppy)₃ into TAZ near the interface, TAZ emission is removed and instead the green portion of the spectra (due to Ir(ppy)₃ emission) is enhanced, confirming formation of excitons on TAZ near the EML/ETL interface and recycling of TAZ excitons for emission. Correspondingly, all type-II devices (with Ir(ppy)₃ concentrations of 0.3–1.0 wt.%) show higher EL efficiencies (maximal quantum efficiencies of 11.5–12%, Table II) than the type-I control device (pure FIrpic device) or all other type-I devices. Among all type-II devices, the device with 0.5 wt.% Ir(ppy)₃ exhibits the highest peak efficiencies of (12%, 35 cd/A, 24 lm/W), although the efficiencies of type-II devices are not very sensitive to the Ir(ppy)₃ concentration in the range of 0.3–1.0 wt.%. The current-voltage-luminescence (*I-V-L*) characteristics of this device are shown in Fig. 9(a),

TABLE II
DEVICE PERFORMANCES OF TYPE-II DEVICES

Ir(ppy) ₃ (wt.%)		0.3	0.4	0.5	0.8	1.0
External Quantum Efficiency (%)	Maximum	11.5	11.6	12.1	11.6	11.4
	@ 100 cd/m ²	11.1	11.5	11.4	11.0	10.9
Luminance Efficiency (cd/A)	Maximum	31.1	33.9	35.3	32.3	33.4
	@ 100 cd/m ²	29.3	31.4	31.4	30.1	30.1
Power Efficiency (lm/W)	Maximum	21.9	22.8	23.9	18.6	18.7
	@ 100 cd/m ²	12.8	13.6	14.0	11.9	11.9
CIE 1931 Coordinates (x, y)		(0.30,0.44)	(0.31,0.44)	(0.31,0.45)	(0.33,0.46)	(0.33,0.47)
Color Rendering Index		58	58	56	60	56
Correlated Color Temperature (K)		6300	6200	6200	5400	5500

Fig. 9. (a) I - V - L characteristics, and (b) external quantum efficiency/power efficiency versus current density for the type-II device with 0.5 wt.% FIrpic in TAZ.

while its external quantum efficiency and power efficiency vs. current are shown in Fig. 9(b). Since the doping in TAZ is low in type-II devices, therefore as in type-I devices, the roll-off in the quantum efficiency with current or brightness remains small over several orders of the current. Even at a brightness of 1000 cd/m², the device retains efficiencies of (11%, 31 cd/A), which are only slightly lower than peak values. Interestingly, we notice that the EL efficiencies (for the forward viewing direction) achieved here are very similar to those recently reported by Forrest *et al.* using blue fluorescence and green/red phosphorescence [23]. Yet, the devices reported here appear to have simpler layer structures, which may have certain advantages in practical applications.

Due to significantly enhanced green emission in the EL spectra, in general the CIE coordinates of type-II devices are

shifted more towards the greenish area, making colors of all type-II devices more greenish white characterized by CIE coordinates of (0.30, 0.44)-(0.33, 0.47) and CCT of 6300–5500 K (Table II). Correspondingly, the CRI's of type-II devices (56–60) are lower than those of type-I devices (62–75). However, one should notice that the composition of the type-II devices is not subjected to thorough optimization yet. For instance, so far only the concentration of the green phosphorescent emitter in TAZ is adjusted, whereas the concentrations of the blue and orange-red emitters (FIrpic, DCJTb) use the values optimized for type-I devices. It is expected that the color performances of the type-II devices can be further improved by optimizing concentrations of all three emitters (FIrpic, DCJTb, Ir(ppy)₃) and the doping regions to shape the spectra. Furthermore, colors may be also improved by using emitters with more saturated colors (e.g., more saturated blue emitters and red emitters etc.).

IV. SUMMARY

In summary, we have investigated white-emitting OLEDs making use of both blue-phosphor-sensitized orange-red fluorescence from the DCJTb dye and the residual blue phosphorescence from the complex FIrpic. By carefully adjusting the concentrations the phosphor and the fluorophore in the emitting layer and choosing the carrier-transport layers (HTL and ETL) for the device structure, WOLEDs containing a single phosphor-sensitized emitting layer (type-I devices) can give colors close to the equal-energy white (0.33, 0.33), CRI up to 75, and efficiencies up to (10%, 23 cd/A, 13.4 lm/W). Further, by doping a green phosphor Ir(ppy)₃ into the poorly emitting electron-transport layer (type-II devices) to recycle excitons formed there, the EL efficiencies can be further enhanced up to (12.1%, 35.3 cd/A, 23.9 lm/W), although the enhanced green emission in the EL spectra makes the color more greenish white and lowers the CRI. In both types of devices, the phosphor sensitization reduces population of triplet excitons in the emitting region and substantially mitigates the efficiency roll-off with the driving current or brightness that is often observed in all-phosphor OLEDs. At the brightness of 1000 cd/m², both types of devices retain quantum and cd/A efficiencies similar to their peak values.

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