Optical characteristics of tandem and microcavity tandem organic light-emitting devices

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Abstract — In pursuit of the further enhancement of the luminance and efficiency of organic light-emitting devices (OLEDs), it is worthy of exploring what benefits could be obtained by combining two luminance-enhancement techniques, i.e., microcavity and tandem OLEDs. Furthermore, a deeper understanding of the optics in tandem OLEDs will be useful for the design and optimization of tandem OLEDs. In this paper, the optical characteristics of noncavity and microcavity tandem OLEDs are theoretically and experimentally investigated. By the use of rigorous electromagnetic modeling of OLEDs, the radiation characteristics of tandem OLEDs as a function of device structures are analyzed and correspondingly, the guidelines for optimizing the performance of tandem devices are suggested. By making use of the analytical results, it is shown that with well-designed microcavity conditions and device structures, a five-fold enhancement in luminance in the normal direction can be achieved with cavity-tandem devices having only two emitting units. A very high efficiency of 200 cd/A for a rather broad brightness range of 100–4000 nits is demonstrated with a phosphorescent cavity two-unit device.

Keywords — Organic light-emitting devices (OLEDs), microcavity, tandem.

1 Introduction
Rapid advances in organic light-emitting devices (OLEDs) and their applications in displays and lighting impose substantial demands in OLED structures having enhanced brightness yet without increasing driving currents.1,2 Two techniques associated with such demands have been developed in parallel in recent years. Tandem OLEDs provide enhanced luminance and current efficiency (cd/A) by stacking multiple emitting units vertically in series.3–5 On the other hand, incorporating well-designed microcavity structures in OLEDs can provide up to a two-fold enhancement in luminance and cd/A efficiency through redistributing the radiation generated in the devices.6–8 In pursuit of the further enhancement of the luminance and efficiency of OLEDs, it is worthy of further exploring what benefits could be obtained by combining both techniques, i.e., by forming microcavity tandem OLEDs. Furthermore, in the few reported tandem OLEDs, one notices that the enhancement in cd/A efficiency with the number of emitting units could significantly exceed a proportional increase as expected from a simple viewpoint.3–5 An understanding of such an unusual efficiency gain and the optics of tandem OLEDs will thus be useful for the design and optimization of tandem OLEDs.

In this paper, we investigate theoretically and experimentally investigate the optical characteristics of noncavity and microcavity tandem OLEDs. By use of rigorous electromagnetic modeling of OLEDs, the optical characteristics of tandem OLEDs as a function of device structures are analyzed, and, correspondingly, the guidelines for optimizing the performance of tandem devices are suggested. By making use of the analysis results, we show that by incorporating the microcavity structure, very high efficiency (200 cd/A) can be achieved with a tandem OLED having only two emitting units.

2 Optical analyses of noncavity and microcavity tandem OLEDs
2.1 Device structures and the optical model used for analyses
Noncavity and microcavity tandem OLEDs used for analyses and in experiments were green phosphorescent OLEDs based on the emitter tris(phenylpyridine)iridium (Ir(ppy)3).9 Also both types of tandem devices employ the doped p-n junction to effectively connect different emitting units.4 The noncavity tandem devices have the typical bottom-emitting structure with a transparent indium tin oxide (ITO) anode on glass substrates and a high-reflectivity Ag cathode [Fig. 1(a)]. The cavity tandem OLEDs adopt the top-emitting configuration with the high-reflectivity Ag as the bottom anode and the semitransparent Ag (20 nm) as the top cathode [Fig. 1(b)]. According to previous studies on microcavity OLEDs,6,8 to maximize the output luminance of cavity OLEDs, a high-reflection and low-loss output mirror is further capped with a 60-nm high-index organic layer 4,4′,4″-tris(N-carbazolyl)-triphenylamine (TCTA, with a refractive index of ~1.77 around 530 nm as measured by ellipsometry) to achieve both high reflection (seen from the inside of the
device) and low absorption at major emission wavelengths of \( \text{Ir(ppy)}_3 \). In the calculation, the distances of an emitting layer (the 15-nm \( \text{Ir(ppy)}_3 \) emitting layer) to both the anode and the cathode are varied to analyze the influences of device structures (e.g., the total thickness of organic layers and the location of the emitter relative to the electrodes, etc.) on the emission and optical characteristics of both noncavity and cavity tandem OLEDs. The use of the organic \( p-n \) junctions for the electrical connection between emitting units allows us to simplify the calculation by using an average refractive index of 1.77 over wavelengths of interest for all organic layers in the devices. The optical properties of Ag and ITO (e.g., reflectivities) used for the analyses are similar to those reported in our previous publications.\(^6,7,14\)

The optical model used for performing the analysis adopts a classical approach based on the equivalence between the emission of a photon due to an electrical dipole transition and the radiation from a classical electrical dipole antenna,\(^10-14\) which can take into account loss due to the electrodes. With the plane-wave expansion of the dipole field, the full-vectorial electromagnetic fields generated by a radiation dipole embedded in a layered structure is calculated, from which the distribution of the radiation power into different plane-wave modes and the far-field radiation related to emission characteristics of an OLED are obtained. In order to model the emission characteristics of an OLED, it is assumed that the emitting layer contains an ensemble of mutually incoherent dipole radiators with distributions in dipole orientations (a random isotropic distribution), locations (throughout the 15-nm emitting layer), and frequencies (using the photoluminescence (PL) spectrum of \( \text{Ir(ppy)}_3 \) as the intrinsic spectral distribution of the dipole radiators). Radiation characteristics of OLEDs are then obtained by averaging contributions over these distributions. In the analyses, the 15-nm emitting layer is freely moved from the electrodes of devices to determine the effects of the emitter locations on radiation characteristics.

### 2.2 Analysis results for noncavity tandem OLEDs

Figure 2 shows the contour plot of the calculated forward luminance resulting from a single emitting layer located at different distances to the cathode and to the anode of the noncavity tandem OLED. The forward luminance is normalized to that of the optimized one-unit bottom-emitting OLED (i.e., the reference device). The dashed lines with a slope of \(-1\) represent the optimized total thicknesses of organic layers (for forward luminance) for one-, two-, three- and four-unit noncavity tandem devices.

![DEVICE STRUCTURES](image)

**FIGURE 1** — Device structures for optical analysis: (a) noncavity tandem device; (b) microcavity tandem device. For one-, two-, three-, and four-unit tandem devices, the total thicknesses of the organic layers between electrodes are set to achieve the first, second, third, and fourth lowest possible resonance modes at 530 nm [near the emission peak of the \( \text{Ir(ppy)}_3 \) emitter], respectively. To simplify calculation, an average refractive index of 1.77 over wavelengths of interest is assumed for all organic layers in the devices. In the analyses, the location of the 15-nm emitting layer is varied in relation to the reflecting back mirror electrode.

![CONTOUR PLOT](image)

**FIGURE 2** — The contour plot of the calculated forward luminance resulting from a single emitting layer located at different distances to the cathode and to the anode of the noncavity tandem OLED. The forward luminance is normalized to that of the optimized one-unit bottom-emitting OLED (i.e., the reference device). The dashed lines with a slope of \(-1\) represent the optimized total thicknesses of organic layers (for forward luminance) for one-, two-, three- and four-unit noncavity tandem devices.
lines happen to pass one, two, three, and four maxima in the contour plot.

For these four tandem devices, their radiation characteristics are further analyzed. Figure 3(a)–3(d) show the calculated forward luminance resulting from a single emitting layer located at different distances to the back mirror electrode in one-, two-, three-, and four-unit noncavity tandem devices, respectively, along with the fraction of normalized radiation power coupled into plasmon modes and waveguided modes (including both substrate and ITO/organic modes). The left and right ends of the horizontal axes in Figs. 3(a)–3(d) represent the organic-metal and organic-ITO interfaces, respectively.

In general, for emitting dipoles closer to the metal, the ratio of substrate modes to waveguided modes are lower, yet most of the radiation power is coupled into the plasmon modes, and the OLED emission is significantly quenched. By increasing the distance of the emitting dipoles from the reflective metal, the radiation power coupling into plasmon modes drops rapidly and those into other modes first rise and then become somewhat oscillatory with distance. The maxima occurring in forward luminance roughly correspond to antinodes of the metal electrodes (i.e., the emitter-to-metal round-trip phase change equals to integral multiples of $2\pi$), while minima correspond to nodes.

Noticeably, emitters located beyond the first antinode of noncavity devices in general contribute a larger forward luminance (up to 1.5 times) than the reference device (i.e., the optimized one-unit bottom-emitting device with emitters located around the first antinode to the metal). Such enhancement is partly associated with enhanced outcoupling of internal radiation because the coupling into plasmon modes has dropped to an almost negligible level and waveguided modes happen to be around their local minima. More detailed analyses also show that the enhanced forward luminance by locating emitters at farther antinodes is also partly attributed to stronger coupling of internal radiation (from emitters at farther antinodes) into the forward direction (i.e., more directed emission as will be shown later in the experiment results).15

The findings here may explain some unusual observations in the recent development of tandem OLEDs.3–5 In the simplest viewpoint, one would expect a proportional increase in the luminance efficiency (i.e., cd/A) with the number of emitting units in tandem devices. In few reported tandem OLEDs, one, however, notices that the enhancement in cd/A efficiency could significantly exceed such a proportional increase.3–5 The results here provide a better understanding of such unusual efficiency gain, since the emitting unit farther away from the reflective metal electrode could contribute a larger cd/A efficiency.

### 2.3 Analysis results for microcavity tandem OLEDs

Similarly, Fig. 4 shows the contour plot of the calculated forward luminance resulting from a single emitting layer located at different distances to the top cathode and to the bottom anode. Again, the forward luminance is normalized to that of the optimized one-unit bottom-emitting OLED (i.e., the reference device), and an array of maxima is observed in the contour plot, where conditions of construc-
tive interference are matched by both electrodes. The four dashed lines with a slope of −1 in Fig. 4 represent the total thicknesses of the organic layers to achieve the four lowest possible resonance modes for a wavelength of 530 nm, which naturally will be conditions for one-, two-, three-, and four-unit microcavity tandem devices.

For these four microcavity tandem devices, their radiation characteristics were further analyzed. Figures 5(a)–5(d) show the calculated forward luminance resulting from a single emitting layer located at different distances to the back mirror electrode (i.e., the bottom-reflecting Ag anode) in one-, two-, three-, and four-unit microcavity tandem devices, respectively, along with the fraction of normalized radiation power coupled into plasmon modes and waveguided modes. The forward luminance is normalized to that of the optimized one-unit bottom-emitting OLED (i.e., the reference device). The left and right ends of the horizontal axes in Figs. 5(a)–5(d) represent the organic-anode (bottom) and organic-cathode (top) interfaces, respectively. In the same tandem device, emitters located around the antinode closest to a metal electrode suffer strong coupling into plasmon modes and thus lower outcoupling and forward luminance. Note that in the microcavity devices, there are two metal electrodes. In contrast, locating emitters at antinodes farther away from both metal electrodes (e.g., the second antinode in the three-unit device and the second/third antinode in the four-unit device) reduces plasmon modes to nearly negligible levels. Although coupling into waveguided modes also increases, yet, in general, stronger forward luminance is obtained.

For microcavity tandem OLEDs, emitters located at all antinodes of each microcavity device in general contribute a larger forward luminance (up to 2.6 times) than the reference device. Such enhancement is partly associated with enhanced outcoupling (considering ratios of plasmon modes and waveguided modes, etc.), such as the case of the
one-unit cavity device (low coupling into waveguided modes). In most cases, however, such enhanced forward luminance is more associated with the stronger coupling of internal radiation into the forward direction (i.e., more directed emission) according to detailed analyses. Such stronger coupling into the forward direction (i.e., more directed emission) results in generally larger contribution (by a factor of around 2) in forward luminance from emitters around an antinode of a cavity device than from emitters around an antinode of a noncavity device.

2.4 Design considerations for tandem OLEDs

Based on results of Figs. 3 and 5, a clear design rule for tandem OLEDs (either noncavity or cavity) is to locate emitters of each emitting unit at corresponding antinodes. Summing the peak values of the forward luminance for each device in Figs. 3 and 5 gives the optimal overall luminance enhancement (relative to the reference one-unit device) that one can obtain for each tandem device. Figure 6(a) shows the luminance enhancement vs. the number of emitting units for both types of tandem devices. For both types of devices, lumiance increases with the number of emitting units, but not linearly. By increasing the number of emitting units (and thus the total thickness of devices), overall coupling (loss) into waveguided modes rises and eventually diminishes the advantages of tandem devices. The deviation is even larger for cavity devices since mode widths (the effective wavelength range of a resonance condition) narrow with cavity lengths.

In tandem devices, not only the brightness but also the operation voltage increases with the number of units. Thus, a fair way to characterize the performance of tandem devices is to evaluate their luminance per (input) electrical wattage. Assuming the operation voltage increases linearly with the number of emitting units, Fig. 6(b) shows such characteristics of tandem devices (normalized to that of the reference device). Noncavity tandem devices exhibit a rather constant enhancement of ~1.2~1.3 vs. the number of emitting units, while an enhancement of ~2.1 and an optimal enhancement of ~2.5 are obtained with one- and two-unit cavity devices, respectively. Thus, in terms of the characteristics of luminance per (input) electrical wattage, two-unit tandem devices (either noncavity or cavity) are the optimized cases. For the optimized noncavity and cavity two-unit devices, one would expect an enhancement of ~2.5 and ~5, respectively, in forward luminance, compared to the noncavity one-unit device. Note that the cavity device gives roughly a further two-fold enhancement due to the microcavity effect.

3 Experiments and discussions

3.1 Device structures for experiments

Experiments were conducted on the reference device (A), noncavity (B), and cavity (C) two-unit tandem devices for comparison with analyses. Device A (Fig. 7) had the structure of glass/ITO (120 nm)/m-MTDATA:F4-TCNQ (2 wt.%, 20 nm)/α-NPD:F4-TCNQ (2 wt.%, 8 nm)/α-NPD (10 nm)/TCTA (10 nm)/TCTA:Ir(ppy)3 (8 wt.%, 5 nm)/TAZ:Ir(ppy)3 (8 wt.%, 10 nm)/TAZ (10 nm)/BPhen (10 nm)/BPhen:Cs (20 mol.%, 27 nm)/Al (1 nm)/Ag (150 nm), which uses conductive doping in carrier-transport layers for current conduction and injection. Organic layers in sequence consist of hole-transport layers (HTL) 4,4’,4”-tris(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA) and N,N’-Di(naphthalen-1-yl)-N,N’-diphenyl-benzidine (α-NPD) with 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4-TCNQ) as p-type conductive doping. Intrinsic hole-transport layers α-NPD and TCTA, double emissive layers (EML) TCTA and 3-(4-Biphenylyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ) doped with Ir(ppy)3, intrinsic electron-transport layers (ETL) TAZ and 4,7-diphenyl-1,10-phenanthroline (Bphen), and BPhen with Cs as n-type conductive doping. Intrinisc organic layers are inserted between emission and conductive transport layers to reduce quenching associated with conductive doping. Based on device A, double-unit tandem devices B and C (Fig. 7) were fabricated with the structures of anode/m-MTDATA:F4-TCNQ (20 nm)/α-NPD:F4-TCNQ (10 nm)/α-NPD (10 nm)/TCTA (10 nm)/TCTA:Ir(ppy)3 (5 nm)/TAZ:Ir(ppy)3 (10 nm)/TAZ (10 nm)/BPhen (10 nm)/BPhen (10 nm)/Al (11 nm)/Ag (150 nm), which uses conductive doping in carrier-transport layers for current conduction and injection. Organic layers in sequence consist of hole-transport layers (HTL) 4,4’,4”-tris(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA) and N,N’-Di(naphthalen-1-yl)-N,N’-diphenyl-benzidine (α-NPD) with 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4-TCNQ) as p-type conductive doping. Intrinsic hole-transport layers α-NPD and TCTA, double emissive layers (EML) TCTA and 3-(4-Biphenylyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ) doped with Ir(ppy)3, intrinsic electron-transport layers (ETL) TAZ and 4,7-diphenyl-1,10-phenanthroline (Bphen), and BPhen with Cs as n-type conductive doping. Intrinisc organic layers are inserted between emission and conductive transport layers to reduce quenching associated with conductive doping. Based on device A, double-unit tandem devices B and C (Fig. 7) were fabricated with the structures of anode/m-MTDATA:F4-TCNQ (20 nm)/α-NPD:F4-TCNQ (10 nm)/α-NPD (10 nm)/TCTA (10 nm)/TCTA:Ir(ppy)3 (5 nm)/TAZ:Ir(ppy)3 (10 nm)/TAZ (10 nm)/BPhen (10 nm)/BPhen (10 nm)/Al (11 nm)/Ag (150 nm), which uses conductive doping in carrier-transport layers for current conduction and injection. Organic layers in sequence consist of hole-transport layers (HTL) 4,4’,4”-tris(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA) and N,N’-Di(naphthalen-1-yl)-N,N’-diphenyl-benzidine (α-NPD) with 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4-TCNQ) as p-type conductive doping. Intrinsic hole-transport layers α-NPD and TCTA, double emissive layers (EML) TCTA and 3-(4-Biphenylyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ) doped with Ir(ppy)3, intrinsic electron-transport layers (ETL) TAZ and 4,7-diphenyl-1,10-phenanthroline (Bphen), and BPhen with Cs as n-type conductive doping. Intrinisc organic layers are inserted between emission and conductive transport layers to reduce quenching associated with conductive doping.
nm)/BPhen:Cs (40 nm)/α-NPD:F4-TCNQ (40 nm)/α-NPD (10 nm)/TCTA (10 nm)/TCTA:Ir(ppy)3 (5 nm)/TAZ:Ir(ppy)3 (10 nm)/TAZ (10 nm)/BPhen (10 nm)/BPhen:Cs (40 nm)/cathode. The noncavity device B used ITO and Al (1 nm)/Ag (150 nm) as the anode and the cathode, respectively, while the cavity device C used Ag (100 nm) and Al (1 nm)/Ag (20 nm)/TCTA (60 nm) as the bottom reflective anode and the semitransparent top cathode. Both tandem devices employed the doped p–n junction to effectively connect two units. The thicknesses of the conductive organic layers were adjusted to match the antinode condition (i.e., locate the emitters around corresponding antinodes) and the resonance condition for the wavelength of 530 nm. The devices were fabricated and characterized as described in Refs. 6, 7, 14, and 15.

### 3.2 Experimental results and discussion

Figure 8 shows the luminance–voltage ($L$–$V$) and the efficiency characteristics of these devices. Reference device A shows steep $L$–$V$ characteristics and low operational voltages typical for OLEDs with conductive doping and $p$–$i$–$n$ structure. Noncavity and cavity tandem devices B and C show operation voltages roughly doubling that of device A, indicating the effectiveness of the conductive $p$–$n$ junction (i.e., BPhen:Cs/α-NPD:F4-TCNQ) in electrically connecting two emitting units. By stacking two emitting units and locating emitters around the antinodes, noncavity tandem device B achieves a maximal efficiency of 105 cd/A, roughly 2.5–2.6 times the device A efficiency (40 cd/A). By incorporating the tandem structure into the cavity, device C shows a maximal efficiency of 200 cd/A, roughly twice the device B efficiency and five times the device A efficiency. It is noticed that a high efficiency of 200 cd/A is obtained at a rather broad brightness range of ~100–4000 nits. In Figs. 6(a) and 6(b), measured efficiency enhancement of devices B and C (solid symbols) are compared with calculated ones (open symbols). Fairly good agreement indicates consistency with the analyses.

Figure 9 compares the electroluminescence (EL) spectrum of the three devices (at a viewing angle of 0° in the forward direction). The EL spectrum of device C is narrowed due to strong microcavity effects, while the EL spectrum of the two-unit tandem device B is similar to that of the one-unit reference device. Figure 10 shows the measured (symbols) and simulated (lines) EL spectra with relative intensities at viewing angles of 0°, 30°, and 60° off the device surface normal for device C. The EL spectra of device
C shows a blue-shift tendency for larger viewing angles due to slightly blue-shifted resonant wavelengths with viewing angles. Since the forward (0°) resonant wavelength of the microcavity device is set near the peak wavelength of the PL spectrum of Ir(ppy)3, the blue-shift of EL with viewing angle is bounded by the short-wavelength falloff of PL. From 0° to 60°, the EL peak wavelength of device C shifts by only ~15 nm. To better quantify differences in colors, CIE 1976 color coordinates (the uniform color space) were calculated from the EL spectra and are shown in the inset of Fig. 10 and Table 1. Such color variations shall be hardly perceptible by the human eye since all the coordinates are closely located at a saturated color coordinate and they differ by at most by ~0.02 on each axis. Figure 11 compares the angular distributions of the measured EL intensity (normalized to the 0° intensity) for the three devices. The conventional single-unit noncavity device A shows the Lambertian distributions. Meanwhile, device B exhibits slightly more directed emission owing to the contributions from emitters at the second antinode to the metal electrode. In the microcavity tandem device C, the more-directed emission is observed due to stronger coupling into the forward direction.

4 Conclusions

In summary, we have investigated theoretically and experimentally the optical characteristics of noncavity and microcavity tandem OLEDs. By use of rigorous electromagnetic modeling of the OLEDs, the radiation characteristics of tandem OLEDs as a function of device structure have been analyzed and, correspondingly, the guidelines for optimizing performances of tandem devices are suggested. Making use of the results of the analysis, we show that with well-designed microcavity conditions and device structures (i.e., consistent with resonant and antinode conditions), a five-fold enhancement in luminance in the normal-direction can be achieved with cavity tandem devices having only two emitting units. A very high efficiency of 200 cd/A has been demonstrated at a luminance of 100–4000 nits with a phosphorescent cavity two-unit device.

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References


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