Transparent organic light-emitting diodes with balanced white emission by minimizing waveguide and surface plasmonic loss

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Abstract: It is challenging in realizing high-performance transparent organic light-emitting diodes (OLEDs) with symmetrical light emission to both sides. Herein, an efficient transparent OLED with highly balanced white emission to both sides is demonstrated by integrating quasi-periodic nanostructures into the organic emitter and the metal-dielectric composite top electrode, which can simultaneously suppressing waveguide and surface plasmonic loss. The power efficiency and external quantum efficiency are raised to 83.5 lm W\(^{-1}\) and 38.8%, respectively, along with a bi-directional luminance ratio of 1.26. The proposed scheme provides a facile route for extending application scope of transparent OLEDs for future transparent displays and lightings.

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1. Introduction

Organic light-emitting diodes (OLEDs) have shown the amazing applications in full-color flat panel displays and solid-state lighting due to their prominent advantages, including low power consumption, light weight, wide color gamut, fast response time and high contrast [1–8]. As a specific device structure, transparent OLED has an additional merit of easy integration with oxide or organic thin-film-transistor driving circuit for active matrix transparent displays with high aperture ratio for potential use in the electronic products, such as smart phones, laptops, automobile windshields, and transparent video screens [9–12]. Therefore, considerable interest has been attracted in the development of transparent OLEDs by introducing new device structures and electrode architectures [9,13–16].

To realize high-performance transparent OLEDs, a major research direction is to develop the alternative transparent electrodes with superior optical and electrical properties for replacing the opaque metal top electrodes that are commonly used in bottom-emission OLEDs. Transparent conductive oxides (TCOs) [11–15,17] and thin metal films [10,18–21] have been extensively studied as transparent top electrodes. For example, indium-tin-oxide (ITO) is one of the most commonly used TCOs in optoelectronic devices because of its unique properties of low sheet resistance (<15 Ω sq−1) and high optical transmittance over the visible region (>90%) [6,15,17,22]. However, deposition of high-quality ITO films as top electrodes in transparent OLEDs requires high processing temperatures, which inevitably causes serious plasma damage to the underlying organic layers due to the bombarding of
highly energetic sputtered particles [20,23–25]. Although various buffer layers have been inserted to reduce the sputtering-induced damage of organic materials prior to the deposition of TCOs, the increased fabrication complexity and degraded device performance hinder the use of TCOs as transparent top electrodes in transparent OLEDs [9,11,15,16,25]. In addition, the use of TCOs as a transparent electrode can also cause optical confinement and total internal reflection of the light generated in organic layers due to the large difference in the refractive indices $n$ among TCOs ($n_{\text{TCO}} \approx 1.8-2.0$), glass substrate ($n_{\text{sub}} \approx 1.5$) and air ($n_{\text{air}} \approx 1.0$). The waveguide loss comprises $\sim$20-30% of internally generated light, limiting the light outcoupling efficiency and severely suppressing top emission in transparent OLEDs [26,27].

Contrast to sputtering-deposited TCOs, thin metal films that hold sufficient electrical conductivity and considerable optical transparency can be thermally deposited at relatively low temperature, which has negligible impact on the underlying organic layers. Therefore, various thin metal films including multilayer stack (e.g., LiF/Al/Ag, Ag/Au) [18,28–32] or alloy forms (e.g., Mg:Ag) [9,25] have been explored as alternative transparent top electrodes in transparent OLEDs. However, a trade-off always exists for thin metal films as transparent electrodes when pursuing both high optical transmittance and low sheet resistance. That is, forming an ultrathin metal film with high optical transparency will sacrifice the electrical conductivity due to its preference for island-like morphology [20,21,33]. Recently, metal-dielectric composite electrodes (MDCEs) have been proposed as promising transparent electrodes regarding optical transparency, electrical conductivity, and large-area film uniformity [34–36]. However, several technical challenges should be overcome when using MDCEs as transparent top electrodes in transparent OLEDs. First, the presence of thin metal films in MDCEs will cause surface plasmonic (SP) loss at the metal-dielectric interface due to the oscillation coupling between free electrons at the metal surface and the emitting dipoles [37–39]. Second, strong optical microcavity effects are accompanied with the use of a flat MDCE structure, giving rise to spectral distortion of angle-dependent emission characteristics and poor color stability [31]. To alleviate the SP loss and the microcavity effects, many efforts have been made by increasing the distance between the emitting layer and the electrode [15,39], inserting a light scattering structure close to the emitting layers [18,40] or roughening the metal-organic interface [41,42]. Among these methods, one potential solution is to implement a photonic nanostructure into the MDCE both for reducing the SP-induced optical loss and for minimizing the microcavity effect with good color quality. Nevertheless, many corrugated structures with a defined period are hindered from practical applications as they often only have positive effects over limited ranges of wavelength and viewing angles. Such a dilemma may be mitigated by introducing a quasi-periodic or quasi-random nanostructure that exhibits broadband spectral dependency over wide viewing angles [4,7]. For example, the MDCE patterned with a quasi-random nanostructure exhibited a low sheet resistance of $\sim$27 $\Omega$ sq$^{-1}$ and an extraordinarily high mean transmittance of $\sim$86% in the entire visible spectral range, which was favorable to flexible OLEDs with broadband and angle-independent outcoupling enhancement when acting as a bottom electrode [36]. Consequently, the integration of the MDCE with outcoupling nanostructures may provide a facial route to effective transparent top electrodes in transparent OLEDs.

In this work, we report an efficient white transparent OLED with an inverted structure by using ITO as bottom cathode and MDCE as top anode, respectively, for highly enhanced and balanced light emission to both sides. The key feature is the combination of the quasi-periodic light outcoupling nanostructure via soft nanoimprint lithography (SNIL) for wavelength/angle-independent white emission. As compared to the conventional transparent OLEDs with a flat device architecture, the suppressions of both waveguide mode and SP losses at ITO and MDCE electrodes are realized simultaneously by integrating the outcoupling nanostructures into organic emitter and MDCE, showing the substantial enhancements of 94% and 58% in light outcoupling efficiencies of top and bottom sides without spectral distortion or angular dependence. The resulting white transparent OLED
yields a bi-directional luminance ratio of 1.26, along with the maximum total current efficiency (CE) of 109.1 cd A\(^{-1}\), power efficiency (PE) of 83.5 lm W\(^{-1}\) and external quantum efficiency (EQE) of 38.8%.

2. Experimental details

Figure 1 illustrates the fabrication process of white transparent OLEDs. ITO-coated glass substrates with a sheet resistance of 20 Ω per square were ultrasonically cleaned with detergent, acetone, ethanol, and deionized water for 20 min in each step and subsequently dried in an oven (110 °C). The ZnO layer was prepared via a sol-gel method with a ZnO precursor solution formed by dissolving zinc acetate dehydrate (Zn(CH\(_3\)COO)\(_2\)·2H\(_2\)O, Sigma-Aldrich, 99.5%, 220 mg, 0.5 M) and ethanolamine (NH\(_2\)CH\(_2\)CH\(_2\)OH, J&K, 60 µL, 0.5 M) in 2-methoxyethanol (CH\(_3\)OCH\(_2\)CH\(_2\)OH, Alfa Aeser, 99.8%, 2 mL) with vigorous stirring for 12 h for the hydrolysis reaction in ambient conditions. The ZnO precursor solution was then spin-coated on cleaned-ITO glass substrate at 4000 rpm for 40 s and then annealed in ambient air at 150 °C for 30 s. The sol-gel-derived ZnO layer was patterned via soft nanoimprint lithography with a polydimethylsiloxane (PDMS) mold having quasi-periodic nanostructures, which was prepared by a series of chemical etching and multitransfer process according to our previous reports [43]. The flexible PDMS mold was covered on the ZnO layer in conformal contact under a constant pressure, and then sintered at 150 °C for 1 min in ambient air. After sintering, the PDMS mold was peeled off, and the substrate was further annealed at 150 °C for 4 min to consummate the high-profile three-dimensional sub-wavelength patterns [44,45]. An about 4 nm thick polyethyleneimine (PEI) layer was spin-coated onto the ZnO layer from a 2-methoxyethanol solution (0.4 wt.%) at 5000 rpm for 50 s, and then was annealed at 100 °C for 10 min in ambient air [46]. Then, the PEI-coated ZnO/ITO-glass substrates were loaded into a high vacuum chamber with base pressure of <2 × 10\(^{-6}\) Torr to complete the device fabrication. The organic emitter and MDCE multi-stack were successively deposited by thermal evaporation with shadow masks without breaking the vacuum, in which the deposition rates and film thicknesses were monitored with quartz crystal oscillators. The effective device area is 0.1 cm\(^2\). For comparison, white transparent OLEDs without and with quasi-periodic nanostructures were simultaneously fabricated in the same batch for the spin-coating process and the subsequent deposition process.

Fig. 1. Schematic illustration of the fabrication process of nanostructured white transparent OLEDs. (a) Spin-coating the ZnO layer on ITO-glass substrate. (b) Imprinting the ZnO layer with the PDMS mold. (c) Demolding the PDMS mold. (d) Spin-coating the PEI interlayer on the patterned ZnO layer. (e) Depositing the organic layers and MDCE multilayer onto the patterned PEI/ZnO layer.
Refractive index ($n$), extinction coefficient ($k$) and film thickness of all the layers were measured using an alpha-SE Spectroscopic Ellipsometer (J.A.Woollam Co., Inc) with the angle of incidence at 70°. Surface morphologies of the quasi-periodic nanostructures were characterized using atomic force microscopy (AFM) (Veeco MultiMode V) in the tapping mode and scanning electron microscopy (SEM) (FEI, Quanta 200FEG). Optical transmission and reflection spectra were recorded with a UV-vis/near-IR spectrophotometer (Perkin Elmer Lambda 750) with an integrating sphere. Current density-voltage-luminance (J-V-L) characteristics and electroluminescence spectra of the white transparent OLEDs were measured simultaneously in ambient conditions using a programmable source meter (Keithley model 2400) and a luminance meter/spectrometer (PhotoResearch PR655). Angle-dependent emission patterns were characterized by placing the devices on a rotating stage with one of the grooves parallel to the rotation axis.

Finite-difference-time-domain (FDTD) method (Lumerical FDTD Solutions 8.7.3) was used for the numerical optical simulation. To calculate the transmittance spectra, a continuous plane-wave source with a broad Gaussian frequency spectrum (300-800 THz) was placed outside a designed 3D unit structure and propagated along the substrate surface normal ($z$ direction), where the boundary conditions of the periodic boundaries in the transverse ($x, y$) directions and the perfect matching layer (PML) boundary in the longitudinal ($z$) direction were employed for the normally incident light source. The electric field radiation distributions of white transparent OLEDs were simulated based on the rigorous electromagnetic theory with one single perpendicular oriented dipole located in the center of the emission layer, using PML boundaries in all dimensions. The dipole orientation was assumed to be isotropic. For simplicity, hexagonal closely packed nanocones with sinusoidal cross-section profile were representatively constructed instead of quasi-periodic nanostructures, where the groove depth, period (400 nm), and fill factor data were set as determined from the AFM images. To calculate the relative extraction efficiency, the emitting excitons were modeled as Gaussian oscillating dipole pulses with a lifetime of 20 fs and a wavelength of 520 nm. The pulse dipole sources with finite and fixed photon number were distributed randomly with an equal number of mutually orthogonal x-, y-, and z-polarizations. In the device simulation design, the film thickness and complex refractive indices of all layers used in the simulation were experimentally determined by an ellipsometer.
3. Results and discussion

Figure 2(a) depicts a schematic diagram of a white transparent OLED constructed with quasi-periodic nanostructures via SNIL technique using PDMS mold (see details in Fig. 1). The device has an inverted structure with ITO and the MDCE multilayer as bottom cathode and top anode, respectively. An 80 nm-thick sol-gel derived ZnO film was firstly spin-coated on ITO-glass as an electron injection layer (EIL). Figure 2(b) is a SEM image of the imprinted ZnO film, revealing that the quasi-periodic nanostructures of the PDMS mold has been successfully transferred to the surface of the ZnO film. Figure 2(c) is an AFM image of the fabricated device, revealing its intrinsic nature of quasi-periodic distribution and sub-wavelength scale. A continuously tapered morphology with a period of ~400 nm and an average groove depth of ~50 nm can be observed on the surface of the transparent OLED, suggesting that the quasi-periodic nanostructures are well retained throughout the successive layer deposition of the device.

As shown in Fig. 2(a), the white organic emitter was constructed with complementary yellow and blue phosphorescent materials, consisting of 1 nm-thick 6 wt.% iridium-(III)bis(4-phenylthieno[3,2-c]pyridinato-N,C2′)acetylacetonate(PO-01)-doped N,N′-dicarbazolyl-3,5-benzene (mCP) layer for yellow emission and 19 nm-thick 8 wt.% of bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl)iridium(III) (Flrpic)-doped mCP layer for blue emission. To match the lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) level of mCP, a 25 nm-thick 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) was used as the electron transporting layer (ETL), and a 45 nm-thick di[4-(N,N-ditolylamino)phenyl]cyclohexane (TAPC) was employed as the hole transporting layer (HTL). In order to reduce the electron injection barrier between the ZnO EIL and the TmPyPB ETL, the ZnO layer was modified by spin-coating a 4 nm-thick PEI interlayer and
then depositing a 10 nm-thick 30 wt.% cesium carbonate (Cs₂CO₃)-doped tris(8-hydroxy-
quinolinato)aluminum (Alq₃) layer prior to TmPyPB deposition [46,47].

The MDCE top anode is in the form of multifunctional stacks, consisting of a
molybdenum oxide (MoO₃) hole-injection layer (HIL), an electrically conductive ultrathin
metal film, and the MoO₃/N,N₀-diphenyl-N,N₀-bis(1-naphthylphenyl)-1,10-biphenyl-4,4₀-
diamine (NPB) optical coupling layer (OCL). As demonstrated previously [48–50], an
ultrathin metal film (≤10 nm) with homogeneous morphology can be achieved by
incorporating a nucleation-seeding layer, reducing both the degradation in optical
transmittance due to metallic particle plasmon absorption and the electrical disconnection due
to isolated granular morphology. To get an ultrathin metal film with high electrical
conductance and low optical loss in the MDCE, a nucleation seeding layer of Al (1 nm), a
pure Ag (1 nm) and a co-deposited Ca:Ag alloy film (weight ratio = 1:1, 8 nm) were
successively deposited by thermal evaporation to solve the dewetting problem and optimize
the film homogeneity [35,36].

In addition to the metallic layer, the dielectric layers play a crucial role in the optical
transmission of a multilayer-stacked MDCE [19]. Therefore, thickness dependence of the
MoO₃ and NPB layers on transmittance of the MoO₃/Al-Ag-Ca:Ag/MoO₃/NPB-structured
MDCE was numerically simulated for light propagation at various wavelengths by using the
FDTD method. As shown in Fig. 3, the optimized layer thicknesses of the MoO₃ HIL and the
MoO₃/NPB OCL are determined to be 20 nm, and 30 nm/40 nm, respectively.
Fig. 4. Optical properties of white transparent OLEDs without and with quasi-periodic nanostructures. (a) Total transmittance of ITO-glass substrate, MDCE structure, flat and nanostructured white transparent OLEDs. (b) Reflectance of flat and nanostructured white transparent OLEDs. (c,d) Photographs of (c) off and (d) on states of nanostructured white transparent OLEDs.

Figures 4(a) and 4(b) display the measured optical characteristics of the ITO-glass substrate, the MDCE multilayer, and transparent OLEDs without and with quasi-periodic nanostructures, which were measured by collecting both specular and diffuse lights using an integrating sphere for the propagation of incident light from the glass side. It is worth noting that the average transmittance of MDCE is ~78.5% over the entire visible wavelength range, showing the good optical transmission for white emission. Correspondingly, the flat device shows a high transparency over the whole visible range with a peak transparency of 70.3% at ~550 nm. Remarkably, nanostructured transparent OLED with quasi-periodic nanostructures exhibits a higher value of transmittance with an enhancement of approximately 8.5% over the entire visible spectrum as compared to that of a flat device. As shown in Fig. 4(a), the peak transmittance is 78.3% around 550 nm and an average transmittance of over 70% is measured over the entire visible regime for nanostructured transparent OLED. It is also noted that the integration of quasi-periodic nanostructures into transparent OLEDs yields a lower reflectance as shown in Fig. 4(b). According to the optical simulation [36], the broadband enhancement on optical transmission in transparent OLED with quasi-periodic nanostructures is attributed to the change of the optical impedance from the flat MDCE multilayer to the nanostructured stack, resulting from the gradient refractive index distribution of the quasi-periodic nanostructures [51–53]. Consequently, the enhanced light transmission can be realized with the reduced reflection loss. Figures 4(c) and 4(d) show the photographs of nanostructured white transparent OLED in off and on states, respectively. The background information can be distinctly observed through the device. Consequently, the superior properties of high transmittance and low reflectance for nanostructured transparent OLEDs match well with the requirements for potential applications in transparent full-color displays and lighting.
Performance of nanostructured transparent OLED was characterized and compared with the flat one. Figure 5(a) shows the J-V-L characteristics of the devices without and with the quasi-periodic nanostructures. Almost identical J-V characteristics for top and bottom emissions indicate similar electrical characteristics in two kinds of transparent OLEDs. The slightly reduced operating voltage for nanostructured device mainly originates from the spontaneously formed corrugated surface as shown in Fig. 1 of AFM and SEM images. It is reported that a stronger electric field distribution will occur due to a partially reduced organic layer thickness in the corrugated structures, resulting in the reduced operating voltage [4,5]. It is obvious that nanostructured transparent OLED shows a distinct improvement in the luminance for both top emission and bottom emission. The device efficiencies are plotted in Figs. 5(b)-5(d) and summarized in Table 1, where the total emission refers to the sum of top and bottom emissions at the same operating voltage. The maximum CE, PE, and EQE of the flat device are 64.3 cd A$^{-1}$, 51.0 lm W$^{-1}$, and 22.8% (38.7 cd A$^{-1}$, 30.8 lm W$^{-1}$, and 13.7% for bottom emission and 25.6 cd A$^{-1}$, 20.2 lm W$^{-1}$, and 9.1% from top emission), respectively. More remarkably, nanostructured transparent OLED exhibits a substantial increase in efficiencies with the integration of quasi-periodic nanostructures. For example, an enhancement factor of 1.72 is obtained for nanostructured transparent OLED, yielding the maximum CE, PE and EQE are increased to 109.1 cd A$^{-1}$, 83.5 lm W$^{-1}$, and 38.8% (60.9 cd A$^{-1}$, 46.8 lm W$^{-1}$, and 21.7% for bottom emission and 48.2 cd A$^{-1}$, 36.7 lm W$^{-1}$, and 17.1% for top emission), respectively. It is noteworthy that nanostructured transparent OLED still maintains relatively high values of 102.3 cd A$^{-1}$, 48.9 lm W$^{-1}$ and 36.5%, respectively, for CE, PE and EQE even at a luminance of 1000 cd m$^{-2}$. Particularly, the more balanced luminance ratio of bottom emission to top emission (1.26:1) is obtained for nanostructured transparent OLED in comparison with the flat device (1.56:1), which is related to the larger efficiency enhancement for top emission than that of bottom emission (1.94 times vs 1.58 times).

Fig. 5. Performance characteristics of white transparent OLEDs without and with quasi-periodic nanostructures. (a) Current density-voltage-luminance (J-V-L) characteristics. (b) Current efficiency, (c) power efficiency and (d) EQE as a function of luminance. Total emission is the sum of bottom and top emissions.
Table 1. Performance characteristics of white transparent OLEDs obtained at the maximum values and at a luminance of 1000 cd m$^{-2}$, respectively. $R_{EQE}$ refers to the EQE enhancement ratio of nanostructured device relative to the flat one at a luminance of 1000 cd m$^{-2}$.

| Device structure | CE(max) | PE(max) | EQE(max) | CE(1000) | PE(1000) | EQE(1000) | $R_{EQE}$ |
|------------------|---------|---------|----------|----------|----------|-----------|----------|
| Flat             | [cd A$^{-1}$] | [lm W$^{-1}$] | [%] | [cd A$^{-1}$] | [lm W$^{-1}$] | [%] | (bottom/top) | (bottom/top) | (bottom/top) | (bottom/top) | (bottom/top) | (bottom/top) | (bottom/top) | (bottom/top) |
|                  | 64.3 | 51.0 | 22.8 | 59.8 | 26.8 | 21.2 | - |
|                  | (38.7/25.6) | (30.8/20.2) | (13.7/9.1) | (36.4/23.4) | (16.7/10.1) | (12.9/8.3) | (−/−) |
| Nanostructured   | 109.1 | 83.5 | 38.8 | 102.3 | 48.9 | 36.5 | 1.72 |
|                  | (60.9/48.2) | (46.8/36.7) | (21.7/17.1) | (57.1/45.2) | (27.6/21.3) | (20.4/16.1) | (1.58/1.94) |

Fig. 6. Angular dependence of emission characteristics of white transparent OLEDs. (a) Normalized EL spectra of white transparent OLEDs obtained at 1000 cd m$^{-2}$ in the direction normal to glass substrate. (b) Normalized angle-dependent emission intensities from bottom and top sides of flat and nanostructured devices. Lambertian emission pattern is displayed as a dashed line. (c,d) CIE color coordinates of (c) bottom and (d) top emissions of flat and nanostructured devices as a function of viewing angle.

Figure 6(a) compares the normalized electroluminescent (EL) spectra of top emission and bottom emission in the normal direction for transparent OLEDs without and with the quasi-periodic nanostructures, exhibiting comparatively symmetric emission spectra from both sides. There is no apparent change in the spectral shape for either top emission or bottom emission of these two devices, indicating the capability of wavelength-independent light extraction of the quasi-periodic nanostructures over the entire visible wavelength range. Figure 6(b) shows the angular dependence of normalized EL intensities of both top emission and bottom emission of transparent OLEDs without and with the quasi-periodic
nanostructures. The angular emission intensities of flat transparent OLED are close to the Lambertian intensity distribution, while nanostructured device shows a stronger side emission. The Commission Internationale d’Eclairage (CIE) color coordinates \((x, y)\) of transparent OLEDs without and with quasi-periodic nanostructures are summarized in Figs. 6(c) and 6(d) for bottom and top emissions, respectively. It is noted that both top and bottom emission spectra of nanostructured transparent OLED remain almost identical with various viewing angles, which are contrary to the flat device showing diverse EL spectra. As viewing angle increases from \(0^\circ\) to \(80^\circ\), changes of CIE coordinates \((\Delta x, \Delta y)\) of bottom emission and top emission from the nanostructured transparent OLED are only \((0.011, 0.009)\) and \((0.014, 0.011)\), respectively, while the corresponding changes of flat device are \((0.031, 0.036)\) and \((0.048, 0.043)\). The superior color stability in the nanostructured transparent OLED indicates the minimized microcavity effect, which can be ascribed to the broadband and quasi-omnidirectional light extraction capability of quasi-periodic nanostructures [4,7,36,42].

To illuminate the light emission behaviors of transparent OLEDs without and with quasi-periodic nanostructures, the near-field light propagation inside the devices was modeled with the FDTD method by locating emitting dipoles of different frequencies with horizontal orientation (parallel to the substrate) in the emitting layer (see the Experimental Section for the detailed procedure of optical modeling). The device structure and complex refractive indices of the materials used for optical modeling were set on the basis of the experimentally determined parameters from the microscopy characterization of layers and spectroscopic ellipsometry. In the optical simulation, hexagonal closely packed nanostructures with continuously tapered profile were used for simplicity instead of quasi-periodically distributed geometry. Figures 7(a) and 7(b) show the \(x-z\)-plane cross-sections of the energy flux density (i.e., the Poynting vector magnitude) from a \(y\)-direction (horizontal) dipole at a wavelength of 520 nm (green emission) in flat and nanostructured devices, respectively. The corresponding simulation results for light propagation at 480 nm (blue emission) and 620 nm (red emission) are shown in Fig. 8. The simulated field intensity distributions show that the quasi-periodic nanostructures have significant influences on the outcoupling enhancements of both top emission and bottom emission of transparent OLEDs. It is worth noting that a substantial amount of radiation is laterally confined at the ITO/organic layer interface (waveguide mode) and organic layer/MDCE interface (SP loss) in the flat transparent OLED (Fig. 7(a)). Contrary to the light confinement in flat device, the radiation trapped in the waveguide mode and SP loss can be effectively released by the quasi-periodic nanostructures in the nanostructured transparent OLED (Fig. 7(b)), resulting in remarkable outcoupling enhancement of radiation propagation through the ITO bottom cathode and the MDCE top anode. Similar behaviors can also be observed for the emitting dipoles at wavelengths of 480 nm and 620 nm (Fig. 8), implying the broadband response to white emission wavelengths. These simulation results are consistent with the strong side emission experimentally observed in Fig. 6(b).
Fig. 7. Normalized cross-section intensity field distributions (at 520 nm) in transparent OLEDs without (a) and with (b) quasi-periodic nanostructures using FDTD method. (c,d) Time dependence of calculated outcoupling efficiency of (c) bottom and top emissions and (d) total emissions from nanostructured device relative to flat device.

Fig. 8. Normalized cross-section intensity field distributions in transparent OLEDs without and with quasi-periodic nanostructures. Propagation of a dipole source at 480 nm in (a) flat and (b) nanostructured devices. Propagation of a dipole source at 620 nm in (c) flat and (d) nanostructured devices.

To quantitatively identify the outcoupling enhancement ratios of top emission and bottom emission, time-integrated photon energy extracted into the air (leaky mode) from both sides
of the transparent OLEDs without and with the quasi-periodic nanostructures were calculated and plotted in Fig. 7(c), which were normalized relative to the saturated extracted energy flow from top emission of the flat device. For both sides of the flat and the nanostructured devices, the time-integrated photon energy extracted into air increases sharply with time and becomes saturated after the complete excitation of the internally generated photons, which are captured by setting transmittance monitors outside the devices. The calculated light outcoupling efficiency of top emission in the nanostructured transparent OLED exhibits an enhancement factor of ~1.93 relative to that of the flat device, while an enhancement factor of ~1.62 is obtained for bottom emission of the nanostructured device. This difference in the enhancement ratio is ascribed to the partial extraction of released photons into the air through the bottom glass substrate. Accordingly, the calculated total light outcoupling efficiency of nanostructured device in Fig. 7(d) shows an enhancement by a factor of ~1.75 compared to that of flat one. It can be seen that the modeling results are in good agreement with the experimental measurements. As reported previously [5], quasi-periodic nanostructure-induced corrugations can provide additional wave vectors for some confined waves (i.e., guided waves and surface plasmonic waves) to satisfy momentum conservation for the outcoupling of confined photons as an extraordinary optical vortex into leaky ( escapable) waves. Consequently, the ITO-related waveguide mode and the plasmonic loss at the MDCE interface in the flat architecture can be effectively minimized by the quasi-periodic nanostructures, yielding the improved light outcoupling and balanced emissions to both sides in nanostructured transparent OLEDs.

4. Conclusion

In summary, high-performance white transparent OLED with almost symmetrical bi-directional light emissions has been demonstrated by integrating quasi-periodic nanostructures for broadband light extraction with little spectral distortion. The transparent device has an inverted structure using ITO as cathode and metal-dielectric composite multilayer as anode, which shows an average transmittance of >70% over the entire visible wavelength range. The maximum power efficiency and external quantum efficiency of the nanostructured white transparent OLED reach 83.5 lm W$^{-1}$ and 38.8%, respectively, with an improvement by a factor of 1.72 as compared with the flat device. Meanwhile, the luminance ratio of bottom emission to top emission is decreased from 1.56 to 1.26, yielding a more balanced bi-directional light emission. According to the optical simulation results, this enhancement on outcoupling efficiency and balanced emission to both sides can be attributed to the extraction of a large amount of internally generated photons trapped in the waveguide and plasmonic modes by integrating the quasi-periodic nanostructures into organic emitter and metal-dielectric composite electrode. In addition to high enhancement in efficiency, the nanostructured device exhibits stable emission color for both top and bottom emissions over the entire visible wavelength range and wide viewing angles. We anticipate that the proposed transparent OLED design provides a promising route for applications in transparent displays and lighting.

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