Research article

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End-emitting nano organic light emitting diodes (OLEDs) with directional output

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Abstract: We report a new strategy for the design of organic light emitting diodes (OLEDs), where nanoscale OLEDs are fabricated into a large-area periodic array with their emission propagating along the active layer and being coupled out through the end facets. A large-area template dielectric grating is produced by interference lithography. The OLED devices are then produced on the side walls of the template grating lines, where each device is carried by the back of a grating line and has a width of <300 nm and a height of about 270 nm. The emission is coupled out of the device on the end facet window after a maximum propagation length of shorter than 300 nm through the active layer, reducing largely metallic absorption by the electrodes and overcoming the optical loss by waveguide confinement. Furthermore, such a configuration enables directional concentration of the output emission. The nanoscale OLEDs also imply large potentials for integration into optoelectronic systems.

Keywords: directional output; end facet output coupling; large-area periodical array; nano OLEDs; organic light emitting diodes.

1 Introduction

Thin-film organic light-emitting diodes (OLEDs) have multifold advantages over their inorganic counterparts, which is mainly based on the molecular materials and the flexible device performances [1–4]. However, due to the

low mobility of the charge carriers, OLEDs have to employ thin-film active layers sandwiched between thin-film electrodes in most of the designs. The main disadvantages of such designs may include the small volume of the active emission medium and the consequently low emission intensity, high optical loss due to the long-distance and long-area interaction of the electroluminescence (EL) with the metallic electrodes [5–7], strong waveguide confinement by the active layer and the transparent electrode, reducing largely the output coupling rate [8, 9].

To overcome above problems, approaches have been investigated with extensive research interests [10–12]. Plasmonic modes related to the metallic electrodes can actually be utilized to mediate the output of the OLED emission. However, extracting more emission confined in the waveguides into the output space is more preferred, since the corresponding light bypath channels constitute the major part of the energy loss of the EL. Constructing structures or patterns on the substrate, which is the output window of the device, has been an important method. Roughening treatment [13], micro-lens-array patterning [14, 15], and micro-/nano-structure patterning [16, 17] of the transparent substrate have been reported to increase the output coupling efficiency of the OLEDs. Alternatively, modifying the internal structure of the device is also an effective approach. This may be achieved by inserting a low-refractive-index or scattering layer between the transparent electrode layer of indium-tin-oxide (ITO) and the substrate to reduce the confinement into the ITO waveguide [18, 19]. Patterning the transparent or the metallic electrodes [20–23] or inserting a photonic layer between the electrode and the substrate [24, 25] has been utilized to enhance the output coupling efficiency. Using high-index substrates or low-index electrodes [26, 27] also increased the extraction of the emission light energy. Additionally, the surface plasmon polariton (SPP) effects can be overcome using transparent electrodes [28]. Furthermore, nanoscale OLEDs have been reported [29, 30], which are important not only for reducing largely the mechanisms for waveguide confinement and the metal-electrode-induced SPP, but also for the integration of the optoelectronic systems. Both the external quantum

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efficiency and the power conversion efficiency may be significantly enhanced [30].

In this work, we report a large-area array of end-emitting nano OLEDs, where we intend to reduce the waveguide confinement effects and the optical absorption by the electrodes. More importantly, the nanoscale metallic electrodes supply strong localized surface plasmons (LSPs) that enhance largely the scattering of light and the output coupling of the device. The end-emitting design also facilitates directional output coupling performance. Conventionally, the directional control of the OLED output has been achieved through patterning the electrodes, the substrates, or the active layers using periodic or photonic structures [31, 32], thus, diffraction of light enabled directional output of the OLED emission. However, the dispersion of the diffraction colors, the low diffraction efficiency, dependence of the diffraction angles and diffraction orders on the grating period may limit the practical application of such designs. In our approach demonstrated here, the directional control of the output is designed to be an intrinsic performance of each OLED. The controllability of the output direction is dependent on the tilting angle of the device, supplying an easy and flexible configuration to overcome the intrinsically existing problems with the OLEDs.

2 Design and fabrication of the nanoscale end-emitting OLED array for directional output

2.1 Design and principles of the nanoscale end-emitting OLEDs

Figure 1(a) demonstrates the design of the OLED array consisting of nanoscale diodes with the output coupled out through the end facet and along the active layer. All of the OLED devices are fabricated on the side walls of the dielectric nanograting lines; therefore, each device is as long as the grating line. However, they have nanoscale dimensions in width and height. This is based on nanoscale dimensions of the grating period and modulation depth. Such a design overcomes the problem of low output-coupling efficiency in conventional designs of OLEDs due to the waveguide confinement and long-distance interaction with the metallic electrodes. In this design, the output of the device is exactly the emission guided by the active layer, so that nearly all emission light energy can be coupled out through both ends after interaction with the metallic electrodes over a distance of shorter than 300 nm.

The periodically arranged nanoscale electrodes also constitute a plasmonic photonic device. Enhanced scattering of light at the end window not only increases further the output coupling efficiency, but also modulates the spectrum of emission into the free space. Optimization of the structural parameters of these nanoelectrodes supplies more space to balance or selectively modulate the EL spectrum.

Apparently, the orientation or tilting angle of the nano OLEDs can be changed by adjusting the slope of the side walls of the grating lines, which can be easily achieved by changing the period, the duty cycle, and the modulation depth of the dielectric grating. Therefore, the direction of the OLED output is also collectively controllable for such a large-scale array. A disadvantage of such a design is that the output is currently through both end facets. However, efficient utilization of the directional output on both sides can be achieved either by utilizing the oppositely directional output for different purposes or by combining them into a single beam. The combination can be realized by producing a high-reflection coating on the top surface of the substrate before the procedures for fabricating the OLEDs.

2.2 Fabrication procedures

Figure 1 (b)–(e) shows the fabrication procedures for the nano OLED arrays on the side walls of a nanograting. Interference lithography was employed to produce the template photoresist (PR) grating with a period of $\Lambda = 500$ nm and a modulation depth of 280 nm, as shown in Figure 1(b). A glass plate with a thickness of 1 mm and an area of 20×20 mm² was used as the substrate. Gold was then deposited onto the side walls of the PR grating lines by oblique evaporation, producing gold nanostrips with a width of about 300 nm, a thickness of 50 nm, and a length of about 10 mm, as shown in Figure 1(c).

Then, PEDOT:PSS and F8BT were spin-coated sequentially onto the top surface of the grating structure. A concentration of 15 mg/mL was used to prepare the F8BT solution in xylene. A spin-coating speed of 2500 rpm and duration of 30 s were employed for the solution-processed deposition of the active layer, as shown in Figure 1(d). The thickness of the F8BT/PEDOT:PSS layer was measured to be about 120 nm. Due to the ~50 nm thickness of the Au nanostrip and the sharp tip of the grating line, the F8BT layer is sheared to be discontinuous at the end of each device, forming the output window of the OLED device. In the final stage, LiF and Al was deposited onto the top surface of the F8BT layer sequentially, where oblique

Figure 1: (a) Schematic illustration of the design of the periodic array of the end-emitting nano OLEDs with directional output. (b)–(e) Fabrication procedures: (b) Interference lithography of a PR-grating template; (c) Oblique evaporation of the Au nanostrips as electrodes on the side walls of the grating lines; (d) Spin-coating of a hole-transport layer of PEDOT:PSS and the active layer of F8BT; (e) Oblique evaporation of the electron-transport layer of LiF and the cathode of Al strips. (f) Cross-sectional SEM images of the end-emitting nano OLEDs.

thermal evaporation at the same tilting angle as that for gold deposition in step (e) was employed. The LiF and Al layers have a thickness of about 1 and 100 nm, respectively, which constitute the cathode of the OLED device, as shown in Figure 1(e).

The thickness of the spin-coated F8BT layer directly affects the performance of OLEDs, which needs to be optimized to achieve the best efficiency of the device. Too thin F8BT layer may result in short circuit of the OLEDs, in

particular near the ridge area of the grating line. However, too thick F8BT layer may lead to large reduction in the modulation depth of the grating; so that nearly a continuous F8BT layer may cover the whole grating and discrete nano OLED devices cannot be achieved. In addition, thick F8BT layer significantly increases the start voltage and reduces the efficiency of the nano OLEDs. We also need to stress that the inclination angle and the gap width of grating lines do not influence the emission efficiency of the

OLEDs, however, these parameters should be designed such that the oblique evaporation of the electrodes and the fabrication of discrete nano OLEDs should be as convenient as possible. Furthermore, the emission from each nano OLED should not be blocked by its neighbors.

Figure 1(f) shows the cross-sectional scanning electron microscope (SEM) image of the final device. A rough measurement on Figure 1(f) gives dimensional values of the nano OLED device, where the OLED device has a width of about 300 nm and a thickness or height of about 270 nm. The width of the device should be a length scale defined by the overlap between the Al and Au nanostrips along the side wall of the grating lines. As has been mentioned above, discontinuity at the edge of the Au strip produced an end facet and supplies an end-emitting output window. This discontinuity of the F8BT layer is dependent not only on the edge steepness of the Au strip, but also on the shape of the top tip of the PR grating lines and the wetting properties of the F8BT/xylene solution on the top surface of the PR grating. However, the SEM image in Figure 1(f) shows a flat end facet, implying all mechanisms favor the formation of high-quality output window at the end of the nano OLED device.

3 Directional output performance of the nano OLEDs with plasmonic modulation

Figure 2(a) shows again the principle design and the structural parameters of the nano OLEDs. The electroluminescence propagates along the F8BT layer after being produced through electrical excitation at different applied voltages. Figure 2(b) shows schematically the experimental design for the measurements on the nano-OLED-array device. Considering that the Au electrode layer is underneath the organic layer, for convenient connection to the direct-current (DC) power supply, a strip of ITO layer was left on the substrate after the etching process, so that the Au film was actually deposited partially on the ITO to act as the anode and the voltage was applied to the device via the ITO layer. The overlap area between the Al and Au strips defines the area of the nano-OLED array, as highlighted by a dashed circle.

Figure 2(c) shows the measurements of the EL spectra at different angles of emission, where the detection head of the spectrometer was oriented at an angle of $\theta = 0 \sim 60^\circ$

Figure 2: (a) Schematic illustration of the nano OLED devices for directional end emitting with a periodical arrangement on the side walls of the grating lines. (b) The structural design and electroluminescence (EL) measurement scheme for the nano OLED array. (c) The EL spectra at different emission angles from 0 to 60 degrees. Inset: definition of the emission angle (θ). (d) Spectral intensity of electroluminescence as a function of the emission angle.

with respect to the normal of the substrate. The definition of the emission angle is as illustrated in the inset of Figure 2(c). The EL spectra are all peaked at about 550 nm and extending from about 500–750 nm. At $\theta = 20^\circ$, an obvious spectral feature is observed at a center wavelength of about 650 nm, which is a selective enhancement and highlighted by a triangle. Clearly, the EL spectrum with a highest peak intensity was measured at $\theta = 30^\circ$, as shown by the plot with red circles, which agrees very well of the tilting angle of the OLED device, as verified by the inset of Figure 2(d). As can be observed in Figure 2(c), when θ is larger than 40° , the emission intensity reduces dramatically. Nearly no EL could be detected at emission angles larger than 60° . The variation of the peak spectral intensity with the emission angle is plotted by the solid red circles in Figure 2(d), which shows convincingly the directional output performance of the device. The range of the emission angles in Figure 2(d) is defined by the divergence of the output, which is determined by the output gap width at the end facet and the diffraction and scattering by the metallic electrodes. This angle range is enlarged due to the far-field measurement, where the detection was carried out at a distance of about 20 cm from the device. Furthermore, the plot in Figure 2(d) exhibits obvious asymmetry, which can be understood by considering that the nano OLEDs are manufactured on one side wall of each grating line, the structures on both sides of the output direction are different. One side is air/Al and the other side is Au/PR. The reflection, scattering, transmission ratios on these interfaces are different. Additionally, the Au electrode on the bottom and the Al on the top have different localized surface plasmons with different absorption and scattering of the emission.

The nanoscale metallic electrodes also supplied strong plasmonic scattering mechanisms for the emission of the OLEDs. The gold nanostrips have a width of about 300 nm and a thickness of about 50 nm, where two localized surface plasmons (LSPs) may be supported by such a nanostructure with the light polarized perpendicular (A) and parallel (B) to the plane of the gold strips, respectively, as defined by the double arrows in Figure 3(a). These LSPR modes can be characterized by the optical extinction spectrum measured on the gold nanostrips by sending a light beam in the plane perpendicular to the substrate at different angles of incidence (θ), which contains the normal of the glass substrate. Obviously, the angle of incidence is defined in the same geometry as the emission angle, as shown in Figure 2(d). Figure 3(b) shows the measured optical extinction spectra of the nano OLED arrays, as the angle of incidence was increased from 0 to 42° . The above mentioned two LSPs can be clearly identified,

which are resonant in the red with a center wavelength tuned from shorter than 600 nm to about 665 nm and in the near infrared with a center wavelength tuned from about 820 to 850 nm. The dependence of the optical extinction spectrum on the angle of incidence is attributed to the different cross-section of the metallic nanostrips facing the incident light beam at different orientations, as depicted by Figure 3(a). These two resonance modes are highlighted by a yellow and red triangle, respectively, in Figure 3(b). It is understandable that the resonance at longer wavelength is already outside the EL spectrum of F8BT, which has nearly no modulation on the output of the nano OLEDs. Therefore, the LSP resonant at shorter wavelengths in the red is responsible for the modulation on the EL spectrum, as shown in Figure 3(c).

In Figure 3(c), we plotted the EL spectra at emission angles of 10° (green), 20° (red), and 30° (blue), as well as that from a conventional F8BT OLED device fabricated on a planar substrate without any structures (solid black curve). For understanding the broadened EL spectra with enhanced red emission in the spectral range from 600 ~ 800 nm for the nano OLEDs, we also included the optical extinction spectrum (dashed black curve) measured on the nano OLED array at an angle of incidence of 30° . Clearly, the LSP centered at about 650 nm is responsible for the spectral modulation in the red, as highlighted by the yellow triangle. The rising edge of the LSP at longer wavelengths has also enhanced the emission at wavelengths longer than 700 nm, however, emission in this band is already very weak and the modulation effect is not obviously identified. The much enhanced optical extinction at wavelengths shorter than 530 nm results mainly from the absorption by F8BT molecules. The analysis in Figure 3(c) verifies the enhanced output coupling of the nano OLEDs by the plasmonic scattering due to the LSP modes in the nanoscale electrodes.

To verify the plasmonic mechanisms more convincingly, we calculated the distribution of optical electric field surrounding a gold nanostrip with a width of 300 nm and a thickness of 50 nm, when it is excited by a light beam from the side and top facets, as shown in the insets of Figure 3(d) and (e), respectively. The polarization of the excitation light beam is shown by the red double arrows in Figure 3(d) and (e). For the excitation along the side end surface at 650 nm, we observe SPP resonance along the gold nanostrip, inducing dipolar electronic oscillation in the direction perpendicular to the top surface. This SPP mode is equivalent to localized surface plasmon on the end facet of the gold nanostrip. In contrast, for the excitation along the normal of the top surface of the gold nanostrip at 820 nm, we can observe a dipolar localized surface plasmon.

Considering that the gold nanostrip is sitting on photoresist and covered with F8BT, which have a similar refractive index of about 1.7, we have used an environmental refractive index of 1.7 in the calculation. Although the simulation results in Figure 3(d) and (e) cannot supply a precise evaluation on the plasmonic response of the whole nano OLED device, these theoretical analysis agree well with the measured spectroscopic response and verify the plasmonic nature of the modulation on the EL spectrum in the red. Thus, the LSP of the gold nanostrips has played a main role in these spectroscopic modulations.

The optical scattering by LSPR is important for increasing the output coupling efficiency and the

brightness of the nano OLEDs. Adjusting the resonance spectrum of the LSPR by changing the structural parameters of the metallic electrodes is helpful for balancing or tuning the light-emitting spectrum. LSPR enables enhanced interactions between the emission, the molecules, and the local field, which supplies new mechanisms for high-efficiency OLEDs. Furthermore, LSPR also supplies mechanisms for polarization control of the output light beam. However, it is also apparent that the LSPR scattering reduces to some extent the long-range directionality of the light emission.

We need to stress here that although the nano OLED devices have a length of millimeters, such waveguides as

Figure 3: (a) Localized surface plasmon resonance of the metallic electrodes of the nano OLEDs and the geometry for the measurements on the angle-resolved spectroscopic response of the nano OLEDs. (b) Measurements of the optical extinction spectra on the nano OLED array at different angles of incidence ($\theta = 0 \sim 42^\circ$) (c) Comparison between the EL spectra of the nano-OLED array at an emission angle of $10^\circ, 20^\circ, 30^\circ$ with that of a conventional F8BT OLED on a planar unstructured substrate. Dashed curve: optical extinction spectrum at $\theta = 30^\circ$. (d) Simulation of the optical electric field in a gold nanostrip with a width of about 300 nm and a thickness of 50 nm for excitation at 650 and 820 nm.

narrow as 300 nm cannot supply strong optical confinement for light propagation at wavelengths longer than 500 nm. Therefore, our structures with a 300 nm width in one dimension actually imply reduction in waveguide losses in two dimensions, as compared with the conventional planar device with a full coverage by a continuous film of the metal electrode. Furthermore, local field enhancement and light-scattering enhancement supplied by the plasmonic interactions, as verified by our spectroscopic characterization in Figure 3, make more contribution to the output coupling than the absorption by the metal electrodes.

4 Optoelectronic performance

Figure 4(a) shows the plots of current density (J) and luminance (L) as a function of the driving voltage (U). A PR-655 SpectraScan/Spectroradiometer from Photo Research Inc. and a Keithley 2400 Source Meter were employed for these characterizations. The measurement results show a turn-on voltage of about 6 V and a maximum current density of more than 150 mA/cm² at a driving voltage of 12 V. However, the device performance declined as the voltage is higher than 11 V, where the peak luminance is about 70 cd/m² at $U = 11$ V and $J = 150$ mA/cm² by direct measurement. This luminance value is still relatively low as compared with those reported for F8BT OLEDs [33–35]. However, we need to consider the following mechanisms for more precise evaluation on the luminance performance: (1) The tilted nano OLED stripe has a width (W) of less than 300 nm and the grating period (Λ) is 500 nm, implying a duty cycle (W/Λ) smaller than 60%. There is thus a factor of at least 1.67 ($\Lambda/W \approx 1.67$) that needs to be taken into account for more precise evaluation and the luminance values are much underestimated by those presented in Figure 4(a). (2) Due to the angle dependence of emission from the nano-OLED array, it is difficult to correct

the values. The most efficient emission is at 30° with respect to the normal of the substrate. However, at such a tilting angle, it is nearly impossible to focus well onto the surface of the device by PR-655. Therefore, the luminance measurements were performed along the normal of the substrate, rather than at an angle of the most intensive emission. According to Figure 2(c), the peak intensity of the EL spectrum at $\lambda_{\max} = 550$ nm along the normal direction is only about 58% that at 30°, or $I_{\theta=0}(\lambda_{\max})/I_{\theta=30^\circ}(\lambda_{\max}) \approx 58\%$, implying another reduction factor of about 1.7, which is evaluated by $I_{\theta=30^\circ}(\lambda_{\max})/I_{\theta=0}(\lambda_{\max}) \approx 1.7$. With the above two factors taken into account, the effective luminance value can be estimated to be higher than 200 cd/cm² for the most efficient directional output. (3) Additionally, the plasmon scattering by the metal electrodes reduced much the collected emission due to the long-range propagation (e. g., over 20 cm) from the device to PR-655, which is another possible reason for the low values of the measured luminance. It needs to be noted that in this work we employed the very basic configuration of conventional OLEDs for each device, where no optimization was made to the electrode or electron/hole transport layers, as compared with those in [36–38], and no improvement was made to the emission collection, as compared with that in [37]. These factors are also account for the measured low efficiency of our device. Nevertheless, our end-emitting nano-OLED array exhibits comparable efficiency as that reported in [38] for a planar F8BT OLED device without TFB interlayer modification.

The inset of Figure 4(a) shows a photograph of the practical device, where we can observe very bright and homogeneous emission within the device area. Figure 4(b) shows the dispersion map with CIE coordinates of the emission from the nano-OLED array, which is within the green-yellow spectrum. The dotted ellipse highlights the directional output performance, verifying the strongest emission is along the direction of about 30° with respect to the normal of the substrate.

Figure 4: (a) Measurements of the J-V/L-V curves. Inset: photograph of the OLED device. (b) The dispersion map with CIE coordinates of the nano OLED array.

5 Conclusions

We report end-emitting OLEDs in nanoscales, which are arranged periodically with controllable orientation angles. Such a design enables directional output coupling of the electroluminescence with high efficiencies. Furthermore, the metallic nanostrips employed as electrodes impose plasmonic scattering of the emission and modulation on the EL spectrum. Such a unique configuration develops a new strategy of OLED designs, which is particularly important for the integration of the OLED devices into optoelectronic systems and for special applications with direct coupling into or excitation of other devices. We need to stress here that the most important reason for the underestimation of the emission intensity of the device is the far-field detection approach. In practical integrated optoelectronics of miniaturized devices, this problem will get overcome automatically, since the emitting light beam will then be utilized in a near-field configuration. More importantly, for near-field application, the smaller divergence of end-facet output configuration, as compared with the conventional planar devices, makes the directionality more obvious and more favorable for the integration of miniaturized devices. Such a design strategy actually enables flexibility in managing the orientation of the device and emission direction of the EL, where the relationship between these two directions can be designed and tuned. Therefore, this design strategy adds a new channel for the development of OLEDs and applies to any thin-film LED devices, which is favorable for miniaturized devices for integrated optoelectronics.

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Author contribution: X. P. Z. designed this research, did the data processing and analysis, and wrote the paper. C. Y. H. fabricated the device and carried out the experiments. Y. W. Z. helped with the device fabrication and luminance measurements.

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