The Influence of the Emission Source on Outcoupling and Directivity of Patterned Perovskite Light-Emitting Diodes

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***Abstract*—The perovskite light-emitting diodes (PeLEDs) have attracted considerable research interests in recent years due to their decent and tunable optoelectronic characteristics. Here, we unveil the properties of the emission source (e.g., location, polarization, etc.) and the influences of the incorporated nanopatterns on the light emission of PeLEDs by using the finite difference time domain (FDTD) method. The results indicated that the PeLEDs with nanopatterns not only show substantial improvement of light extraction intensity but also reveal a remarkable directivity. Moreover, the emission angle of the directional PeLEDs can be continuously engineered over 65º by judicious selection of the nanopatterns. This work is of great importance for engineering highly directional and efficient luminescence devices.**

***Index Terms*—Perovskite light emitting diodes (PeLEDs), directivity, light extraction, nanopatterns.**

I. Introduction

METAL halide perovskite light-emitting diodes (PeLEDs) reveal the advantages over organic light-emitting diodes (OLEDs) due to their outstanding optoelectronic characteristics, such as excellent color purity [1], high brightness [2], and tunable luminescence [3]–[5] etc., which are very suitable for the promising applications covering the general lighting to next-generation flat-panel displays [6]–[7]. There has been considerable amount of research and development for the improvement of efficiency through incorporation of various nanostructures into PeLEDs, for instance, the nanopatterns on transparent substrate (destroy substrate mode [8]–[9]), surface texture and roughness (utilize diffraction effect [10]–[12]), metallic nanostructures (enhance light-matter interaction by plasmonic effect [13]–[15]) and photonic crystals (manipulate light by Bloch mode [16]). The reported external quantum efficiencies of red, green and blue PeLEDs have substantially improved to 20%, 20%, 10%, respectively [17]. Nevertheless, particularly worth mentioning is that the approaches to achieving the directional PeLED have rarely been studied. It is also an essential topic to manipulate the radiation and directivity of the lighting devices, which has various application prospects in daily life such as visible light wireless communication [18], biological sensors [19] and autostereoscopic naked-eye 3D displays [20]–[21] etc. The typical strategies to achieve directional emission include, e.g., embedment of scatters at the top of light emitting devices [22], adoption of optical cavities such as the distributed Bragg reflector (DBR) [23] and photonic crystals into the organic layers [24]. The above methods enable the directional emission at a specific radiation angle, while there are few works on achieving the continuous modulation of the light emission angle. Therefore, it is desirable to offer a feasible approach, which can not only enhance the PeLED radiation efficiency but also control the directivity of the outcoupled light.

In this work, various nanopatterns have been applied into electron transport layer (ETL) of planar PeLED to improve the light extraction efficiency and achieve high directionality of emission. Firstly, we study the characteristics of the emitting source on the light extraction efficiency by analyzing the possible optical loss channels in a planar and dielectric patterned PeLED, respectively. Meanwhile, a directional PeLED can be obtained by appropriately selecting nanopatterns. Subsequently, the continuous modulations of the emission directivity have demonstrated for the PeLEDs through judiciously tuning the periodicity of the nanopatterns. Finally, we also have found that the intensity of PeLED with metallic nanopatterns is obviously enhanced, and the PeLED with hybrid (dielectric and metal) nanopatterns can stimulate more emission angles. Our work will be very useful in improving the efficiency of PeLEDs and contribute to the further development of directional PeLEDs and other lighting applications.

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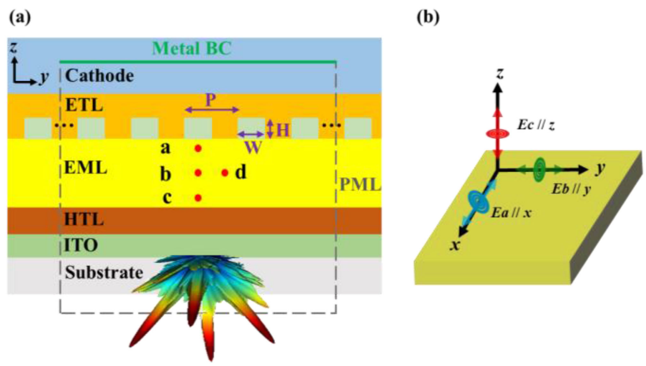

Fig. 1. (a) Device structure of perovskite light-emitting diodes with the incorporation of nanopatterns, the geometrical parameters of the nanopattern is $P = 400$ nm, $W = 200$ nm, $H = 40$ nm, duty cycle is 0.5. The metal boundary condition (BC) is imposed on cathode, and perfectly matched layer (PML) has applied for other boundaries. EML represents the emission layer, HTL/ETL denotes hole/electron transport layer. (b) Schematic illustration of the dipole polarization.

II. Device Structure and Simulation

The device structure of the proposed PeLEDs is schematically shown in Fig. 1(a). The stacked PeLEDs consist of the substrate glass, indium tin oxide (ITO) anode, hole transport layer (HTL), emission layer (EML), electron transport layer (ETL), and aluminum cathode. The commonly used methylammonium lead triiodide (MAPbI3), poly(34-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT: PSS), an [6, 6]-phenyl C61 butyric acid methyl ester (PCBM) has adopted as the emission layer, HTL and ETL, respectively. The periodic dielectric/metallic nanopatterns will be embedded into the ETL of PeLEDs. The refractive indices of the corresponding materials are summarized in Table S1 in Supporting Information. The effect of Fresnel reflection[25] between air and glass will be considered with the finite difference time domain (FDTD) simulation method. We adopt a classical electrical dipole model [26] to simulate the incoherent spontaneous emission process. Moreover, dipole emitter that highly dependent on the surrounding environment will be fixed at four representative locations (denoted as red dot in Fig. 1(a)), which indicate the light emission from bottom/top surface and bulk region of perovskite material. The metal boundary condition was used in the cathode and the other directions adopt perfectly matched layers to simulate the infinite space. The far-field radiation intensity was obtained by collecting the near field data at horizontal direction and then adopting the near-to-far field transformation. In addition, to describe the incoherent and random process of the emitted photons inside perovskite material, the expression [27]: $|\boldsymbol{E}|^2 = (|\boldsymbol{E}_a|^2 + |\boldsymbol{E}_b|^2 + |\boldsymbol{E}_c|^2)/3$ represents the average electric field intensity from an ensemble of incoherent and isotropic dipole emitters. Here, $|\boldsymbol{E}_a|$, $|\boldsymbol{E}_b|$ and $|\boldsymbol{E}_c|$ denote the electric field intensity generated by the dipole emitter with polarization along the x-, y- (i.e., horizontal) and z- (perpendicular) axis, respectively. The light extraction efficiency (LEE, $LEE = P_{out}/P_{source}$) is defined as the ratio of the power flux extracted from the structure with respect to the overall emitted power from the dipole emitter source [28]. The polarization properties of the emission source on the LEE has also been considered, the dipole source with perpendicular (i.e., z-direction) and horizontal (x-direction) polarization schematically depicted in Fig. 1(b) are considered in the simulation.

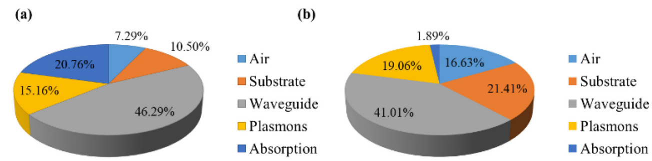

Fig. 2. Proportion of optical loss power of (a) planar and (b) nanopatterned PeLED at the wavelength of 780 nm, respectively.

III. Results and Discussion

A. Radiation Pattern Analysis of Vertical Dipole Location

The total optical power dissipation proportion has obtained for the planar and dielectric (such as ZnO) patterned PeLED. The existing optical loss is mainly ascribed to the air mode, substrate mode, waveguide mode, surface plasmon polariton (SPP) mode induced by the metal electrode, and absorption loss [29]. The substrate mode is resulted from total internal reflection (TIR) [30] between air/substrate interface, the waveguide mode refers to a large amount of photons, which are trapped in the active layer and ITO anode due to the mismatched refractive indices of the stacked layer. The SPP mode at interface between dielectric and metal is derived from resonance between collective oscillations of electrons in metal and the electromagnetic waves[25]. The waveguide mode loss is about 46.29%, which plays a primary role in the planar PeLED due to its high index. On the other hand, the SPP mode loss, about 15%, as a form of non-radiation loss also occupied a large proportion except absorption. Recent studies have demonstrated that the efficiency can be improved for the device with nanopatterns due to the extraction of trapped light that previously falls into the escape cone. As shown in Fig. 2, the light extraction from the device into the air can be enhanced over two times compared with flat structure. The in-depth reason is the reduction of photons previously dissipated as absorption loss, which now is out coupled into air. Interestingly, the incorporation of nanopatterns between the active layer and transparent electrodes will also couple the waveguide modes to substrate modes.

Meanwhile, the far-field intensity of multi-layered planar PeLEDs has been extracted and the dipole emitters are located in the perovskite layer along the vertical direction (i.e., point a, b, c in Fig. 1(a)) to investigate the effect of optical cavity on the angle of emitted light. As shown in Fig. 3(a), the strongest light intensity has observed for the dipole located at the top surface (i.e., position a, near the back opaque cathode), which shows a typical Lambertian radiation pattern [31] indicating the even distribution of the emitted photons in all directions. When the dipole emitter moves to the bottom surface (i.e., location c, near front transparent anode), the light intensity becomes weak and the far-field radiation pattern turns into the dumbbell pattern (green dash line in Fig. 3(a)) following the radiation pattern

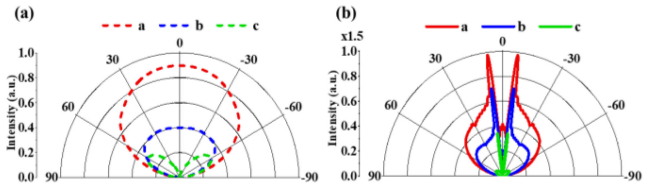

Fig. 3. The radiation pattern of the dipole located at three vertical positions (a) planar PeLED and (b) dielectric patterned PeLED.

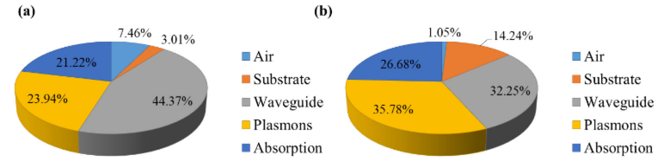

Fig. 4. Proportion of optical loss power of (a) horizontal and (b) perpendicular polarization mode of planar PeLED at the wavelength of 780 nm, respectively.

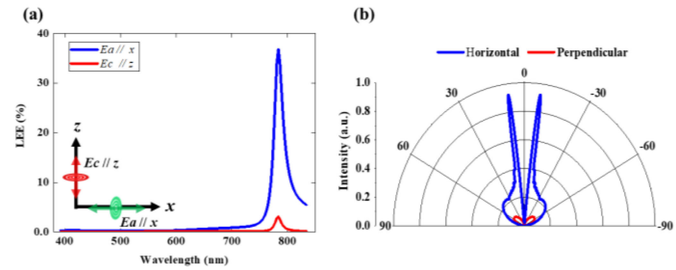

Fig. 5. (a) The light extraction efficiency and (b) radiation pattern of dielectric patterned PeLED with dipole polarization mode of horizontal and perpendicular, respectively.

of the dipole emitter. In order to study the reason of emission pattern formation as well as the emission intensity enhancement, we calculated the Purcell factor which is defined as the ratio of the dipole emission power in the PeLED device to that power in an infinite homogenous medium [32]. Purcell factor shows enhancement in the spontaneous emission rate of an emitter inside a microcavity. The dipole emitter located near the cathode (position a) will induce the highest enhancement of spontaneous emission rate (see Fig. S1 in Supporting Information) as compared to the other two points (i.e., point b and c). Hence, the light emission for the dipole source located at point a will present the strongest intensity due to the boosted spontaneous emission rate. Moreover, the waveguide mode loss is the major limitation on the LEE because the presence of the TIR effect will form a narrow escape cone for the emitted photons particularly for high index perovskite. The escape cone at the two interface for the emitted photon can be estimated by the critical angle $\theta_{\text{critical}} = \arcsin (n_1/n_2)$, in which n_1 and n_2 represent the refractive index of two interfacial materials, respectively. For instance, the critical angle θ_{critical} at the interface of the perovskite/PEDOT: PSS ($n_{\text{perovskite}} \approx 2.57$, $n_{\text{PEDOT:PSS}} \approx 1.45$ at the wavelength of 780 nm) and glass/air are around 33.9$^{\text{o}}$ and 41.8$^{\text{o}}$, respectively. When the dipole emitter near the front transparent anode, the incident angle of the emitted photons will be changed, and only a small amount of photons falls into the escape cone and then goes outside the device into air, a large amount of photons beyond the critical angle will be trapped in the perovskite and carrier transport layer due to TIR effect forming the waveguide modes, which subsequently will be absorbed by the lossy material and turned into heat dissipation.

In fact, due to the presence of the TIR effect, the unidirectional photon emission is disadvantageous for light outcoupling and subsequently limits the LEE of multi-layered PeLEDs. Regarding the dielectric patterned PeLEDs, the highest Purcell factor occurs at the working wavelength of 780 nm indicating the strong spontaneous emission rate at the emission wavelength (Fig. S2a). The Purcell factor together with the corresponding far-field radiation pattern (Fig. S2b) of dielectric patterned PeLED further confirm the outcoupling enhancement by the diffraction effect and Purcell effect. The underlying reason is that more photons will fall into the escape cone due to the diffraction effect of dielectric nanopattern, which then induce the directional emission pattern. Together with the enhancement of spontaneous emission rate at the working wavelength 780 nm, the waveguide mode loss will be reduced. Therefore, the far-field intensity of the patterned PeLEDs has enhanced by 1.5 times as compared to planar PeLEDs (Fig. 3(b)) Interestingly, the far-field pattern of the emitted light shows the distinct directivity and the maximum value achieved at angle of $\sim$10$^{\text{o}}$.

B. Light Extraction for Polarized Dipole

Instead of extracting the energy from the device by using nanopatterns, the dipole polarization characteristics will also affect the light extraction efficiency and the directivity of emitted light. The fraction of optical loss of horizontal and perpendicular polarization mode of dipole emitter have considered for the planar PeLED (Fig. 4). Meanwhile, the light extraction efficiency of horizontal (x- direction) and perpendicular (z-direction) polarization of dipole in nanopatterned PeLED has considered, since there is no anisotropy in the *xy* plane [33]. Two type polarizations of dipole emitter are depicted in the inset of Fig. 5(a). For the planar structure, the power outcoupled into the air for the emission source with horizontal polarization mode (Fig. 4(a)) is slightly better compared to the one with random polarization mode (Fig. 2(a)). However, emitting the power into the air for the completely perpendicular polarization mode of the dipole is extremely less due to the higher SPP mode loss, particularly in nanopatterned PeLED. As shown in Fig. 5, the light extraction intensity for the dipole emitter with horizontal polarization dramatically outperforms the one with perpendicular polarization. The main reason is that the emitted pattern of the horizontal polarized dipole is vertical to the perovskite layers and a large amount of emitted photons falls into the escape cone and then escape into the air. When the dipole emitter is perpendicularly polarized, the emitted pattern of the light is parallel to perovskite layer and a large amount of the emitted photons is trapped in the perovskite layers in terms of waveguide mode and the substrate. Therefore, the dipole emitter with the horizontal polarization reveal better light extraction

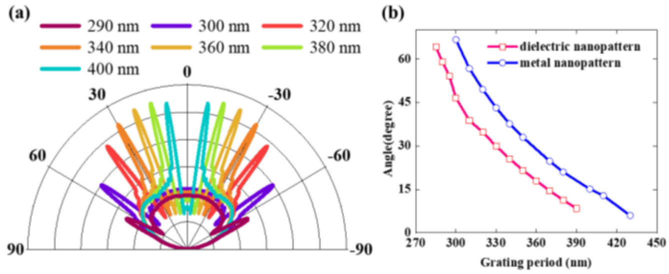

Fig. 6. (a) Radiation pattern of a PeLED with different dielectric nanopattern period. (b) curves of emitted angles with dielectric and metal nanopatterns period, respectively.

efficiency compared with the perpendicularly polarized dipole. The engineering of isotropic emitting source into polarized one will be an alternative approach for light extraction enhancement.

C. Continuous Modulation of Emission Angle

The outstanding emission directivity can be observed through introducing dielectric nanopatterns into the device. To further understand the influence of the nanopattern geometrical parameters on the emission angle, we examine the modulation of the nanopattern periodicities from 290 nm to 400 nm (with fixed duty cycle of 0.5). The emission angle of the directional PeLEDs can be manipulated from 5º to 65° for both dielectric and metal nanopatterns (Fig. 6). When we increase the period of the nanopattern, the emission angle of the photons will be altered from 65º to 0º due to the improvement of zero-order diffraction mode. Therefore, the judicious selection of the incorporation nanopattern will provide us a sensible method to achieve continuous modulation of directional PeLEDs.

D. Radiation Pattern Analysis of Metal and Hybrid Nanopatterned PeLED

Regarding the improved light extraction efficiency by the diffraction effect of the incorporated dielectric nanopattern, the metallic nanopattern (such as Ag) possessing the plasmonic effect has been used to further boost the intensity of the light emission. As shown in Fig. 7(a), the far-field intensity has substantially increased by 8 times for the PeLEDs with Ag nanopatterns as compared to the planar one, which is mainly due to the synergetic effect from the diffraction and plasmonic effect of Ag nanostructures. We also calculated the near-field pattern of metal and hybrid patterned device structure. The results shown in Figs. 7(c)–(d) indicated that the metal patterned PeLED has the considerable near-field enhancement mainly relying on the plasmonic effect. At the same time, it's well known that dipole emitter placed in a strong electromagnetic field environment will have a higher Purcell factor. Thus, the strong light-matter interactions induced by Ag nanopatterns also improve the PeLED directivity and increase the maximum emission angle from 10º to 15° compared with dielectric nanopatterned PeLEDs (Fig. 3(b)). In addition, we also examine the variation of the dipole location along the horizontal direction (i.e., point b and d in Fig. 1(a)) on the light extraction efficiency. The LEE enhancement will be

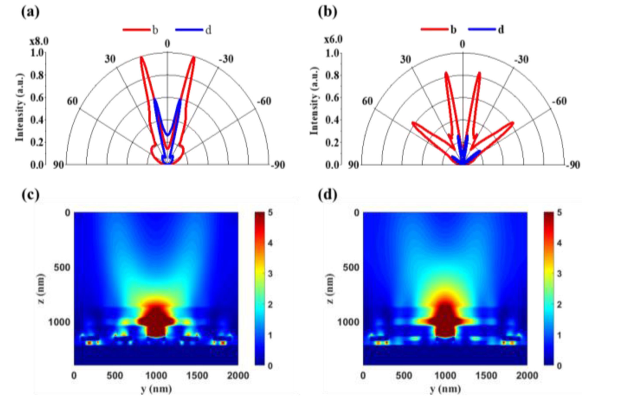

Fig. 7. The far-field radiation pattern of (a) metal and (b) hybrid patterned PeLED. The near field distribution of (c) metal and (d) hybrid patterned PeLEDs.

slightly weakened as the dipole emitter away from Ag nanopatterns, which may due to the reduction of the coupling strength between the dipole and metal nanopatterns, and then shift of the dominant diffraction from low order to high order resulting in more photons beyond of the critical angle [34].

In spite of the LEE enhancement from both dielectric and metallic nanopatterns, it is also appealing to study the PeLEDs with the hybridized (such as ZnO and Ag) nanopatterns. Interestingly, the adoption of hybridized structure with alternatively repeated ZnO and Ag nanopattern enables to produce an additionally distinct emission angle around 60º (Figs. 7(b)–(d)). The existence of mode coupling between dielectric and metallic nanopatterns will induce the energy transfer from the low-order mode to the high-order mode, which will slightly weaken the maximum light intensity. Moreover, we have also found that the manipulation of the dipole emitter location in between ZnO and Ag nanopatterns will alter the radiation pattern and reveal the asymmetric properties.

IV. Conclusion

In summary, we analyse the fractions of various optical loss mode under different emission source characteristics and propose the incorporation of dielectric and metallic nanopatterns as well as their hybridized nanopatterns to achieve the tunable directional PeLEDs. We have unveiled the continuous modulation of the incorporated nanopattern enables to substantially modify the emission angle. The directivity of PeLEDs can be remarkably tuned from angle of 5º to 65º. Besides, we also found that the dipole with the horizontal polarization is more beneficial to light extraction. Our work offers a flexible approach to realize the controllable directional emission of PeLEDs and opens the feasible way for controlling the spontaneous emission of PeLEDs.

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