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Design of a Horizontally Aligned Perovskite Nanowire LED With Improved Light Extraction

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ABSTRACT Within a short span of a few years, metal halide perovskite light-emitting diodes (LEDs) have shot past the 20 percent external quantum efficiency mark. As the material quality and photoluminescent quantum yield of perovskite are already at par with the state-of-the-art, light extraction for further quantum efficiency enhancement has taken the center stage of scientific interest. Herein, we demonstrate horizontally aligned perovskite nanowire-based light-emitting diodes with 50.9 percent of light extraction efficiency, with a 2.9-fold increase compared to the planar counterpart. The absorption spectra of the plane wave incidence exhibit the weak trapping of light within the emission range. Purcell factors of horizontal perovskite nanowires with different diameters are also studied, which provides design guidelines for applying the horizontally aligned perovskite nanowires for multifarious optoelectronic applications such as lasing. The resonance peak positioned at 530 nm wavelength shows the nanowire antennas' effect in the nanowires with diameters of 300 nm, 500 nm, and 550 nm respectively. Furthermore, the fabrication process of the light-emitting devices based on ink-jet printing has also been proposed. This report not only provides an in-depth understanding of light extraction in nanowire LEDs but also heralds the inception of horizontally aligned perovskite nanowire-based optoelectronic devices.

INDEX TERMS Perovskite, light-emitting diodes, light extraction, Purcell factors.

I. INTRODUCTION

Light-emitting diodes (LEDs), such as gallium nitride (GaN) LEDs, organic LEDs (OLEDs), and metal halide perovskite (PRK) LEDs, have become instrumental and promising building blocks in displays and lighting [1]–[4]. Reported for the first time in 2014, PRK green/red/infrared LEDs have come a long way in surpassing 20% external quantum efficiency (EQE) which happens to be one of the most important figure-of-merits for LEDs [5], [6]. High material quality usually leads to a high radiative recombination rate and subsequently high photoluminescent quantum yield (PLQY) [7]. As the PLQY of PRK materials has touched almost unity, further EQE enhancement relies more on sufficient light extraction efficiency (LEE) [8]. PRK LEDs with planar structure have large total internal reflection which is attributed to the high refractive index of the perovskite. However on the other hand, the nanowire structure potentially reduces the total internal reflection. In this work, we discuss a design of horizontally aligned PRK nanowire (PRK NW) LED that possesses the LEE up to 50.9%, with 2.9-fold enhancement compared to the planar counterpart.

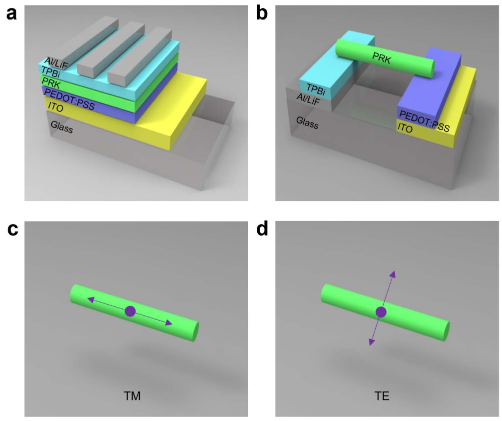

FIGURE 1. Device structures of (a) the planar PRK LED and (b) the horizontal PRK NW LED. Dipole source polariztion directions for (c) TM and (d) TE for LEE simulaitons. The purple arrow is the E field polarization direction.

TABLE 1. Calculated *LEE* for horizontal NWs with different diameters.

Diameter (nm)	LEE_{TE} (%)	LEE_{TM} (%)	$LEE_{\text{weighted average}}$(%)
100	20.6	61.2	34.1
200	28.8	54.7	37.4
300	25.5	60.7	37.2
400	34.7	58.1	42.5
450	36.6	60.5	44.6
500	40.7	66.6	49.3
550	45.2	62.4	50.9
560	42.1	60.3	48.2
580	40	57.3	45.8
600	31	49.2	37.1

II. RESULTS AND DISCUSSIONS

As an alternative of the traditional planar glass/ITO/PEDOT:PSS/PRK/TPBi/LiF/Al (ITO, indium tin oxide; PEDOT:PSS, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; TPBi, 2,2',2"-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole); LiF, lithium fluoride; Al, aluminum) structure (Fig. 1a), the horizontal PRK NW is used as the active layer, as shown in Fig. 1b. Light is emitted from the PRK NW.

ITO and Al/LiF are both 200 nm thick. LiF, which works as the interfacial layer between the cathode (Al) and electron injection layer (TPBi) is around 2 nm thick. Both TPBi and PEDOT:PSS have a thickness of 40 nm. The length of PRK NW is fixed to 2 μm. The diameter of PRK NW is systematically tuned from 100 nm to 600 nm for methodical optimization. Refractive indices of PRK/TPBi/PEDOT:PSS are 2.2/1.7/1.5, [9] and the indices of Al and ITO can be obtained from the online database [10]. The PRK NW can be fabricated by mixing PRK with PEO (PEO, poly(ethylene oxide)). As PEO has a refractive index of ~1.5, the mixed PRK/PEO resulted in a refractive index smaller than 2.2. A smaller index mismatch between the active material and the surrounding environment potentially reduces the total internal reflection while engendering the LEE at the same time.

As can be seen in Table 1, LEEs have been calculated for different PRK NW diameters. Interestingly, LEEs are also related to source polarizations. Dipole sources with transverse magnetic (TM) mode parallel to the PRK NW and transverse electric (TE) mode perpendicular to the PRK NW are shown in Fig. 1c&1d.

In our simulation, both top emission (P_{top}) and bottom emission (P_{bottom}) are included as the output power. As for the planar device structure, the common metal electrode on either top or bottom works as the reflector and dissipates the surface plasmon polariton mode. The source power is normalized to 1. The LEE is calculated by the formula:

$$LEE = \frac{\left(P_{\mathrm{top}} + P_{\mathrm{bottom}}\right)}{P_{\mathrm{source}}} \tag{1}$$

As the NW is axially symmetric, the final LEE for NW is

$$LEE_{\text{weighted average}} = \frac{2}{3}LEE_{\mathrm{TE}} + \frac{1}{3}LEE_{\mathrm{TM}} \tag{2}$$

We calculate the LEE for different NWs with diameters increasing from 100 nm to 600 nm (Table 1). Intriguingly, the highest average LEE is 50.9% at the optimized diameter of 550 nm. Meanwhile, TM dipole always has better LEE than TE dipole, which is consistent with Grzela *et al.*'s report [11].

For a comparison, we simulate the LEE of the planar structure device with the same thicknesses of each layer. The average *LEE* is found to be 17.5%.

It's worth noting that, the length of the NW can impact the light extraction, as well as the carrier transport and recombination within the PRK. Considering that most of the state-of-art PRK LEDs have a very thin (tens of nanometers) active layer, the optimal NW length can be varied for different scenarios. Simultaneously, we should also note that the carrier diffusion length can even reach ~10 μm level in monocrystalline PRK materials [12]–[15].

Besides the light out-coupling, carrier injections and radiative recombination rates also significantly affect the EQE of LED devices [8]. To improve the EQE for real devices, both the electrical and optical parameters need to be taken into consideration and well-optimized. In nanostructured LEDs, the significantly increased LEE can notably improve the EQE. However, it requires more sophisticated fabrication techniques compared to the planar architecture, which in turn may introduce defects and thereby affect the purity of the material system.

As a result, the EQE can be reduced if the fabrication processes of the horizontal nanowire LED devices are not well designed, optimized, and simplified. The length of 2 μm may not impact carrier transport in the device with high quality NWs. Moreover, the NW length also needs to be optimized in terms of both electrical and optical factors.

In the absorption spectra simulation (Fig. 2), the plane wave with the wavelength ranging from 400 nm to 600 nm

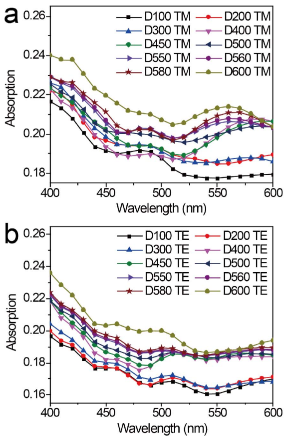

FIGURE 2. Absorption spectra of normally incident TM and TE plane waves for NWs with different diameters.

TABLE 2. Absorption peaks for NWs with different diameters (all the peaks here are weak absorption peaks).

Diameter (nm)	Absorption peaks for TE (nm)	Absorption peaks for TM (nm)
100	510	480
200	510	n.a.
300	510	480, 580
400	510	490, 590
450	510	490, 590
500	510, 585	n.a.
550	500, 590	485, 560
560	500	485, 560
580	500	490, 570
600	460, 500	560

is normally applied onto the PRK NW device (device structure shown in Fig. 1). We assume real constant indices of PRK, PEDOT:PSS, and TPBi for all the wavelengths (400-600 nm). The E fields for TM and TE are parallel to the axial and radial directions, respectively. The light absorption of NWs with different diameters and different polarizations have been provided in Table 2.

The 530 nm emission wavelength corresponds to green PRK LED materials such as cesium lead bromide ($CsPbBr_3$). The absence of strong absorption peaks at 530 nm indicates weak trapping of the light, which results in a high LEE. It's worth noting that usually light with grazing incident angles is likely to excite guided/leaky modes, and light with the normal incident angles can excite Mie resonance for infinite long NW [16]. Our NW has a finite length, but the normal incident light does not excite clear guided/leaky modes, as can be confirmed from the absorption spectra.

In Fig. 3, we simulate the cross-sectional E^2 field profiles at the center of the NW as an indication of light behavior in the horizontally aligned PRK NWs. No strong resonance in the NW is observed, which is consistent with the absence of a strong resonance peak in the absorption spectra. If a

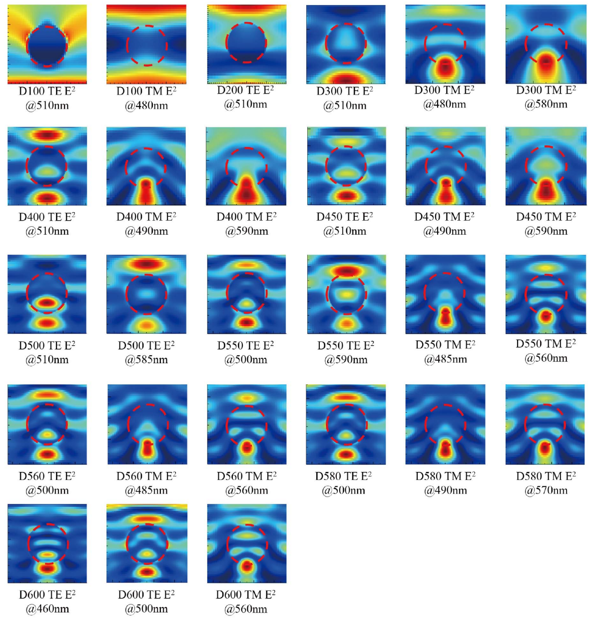

FIGURE 3. E^2 field profiles at the center of the PRK NWs for TE and TM light sources (plane wave) at different absorption peaks.

strong resonance peak exists, the generated light will be trapped inside the NW as the guided modes, which is not favorable for light-emitting purposes. Intriguingly, TE and TM incidences can excite whispering-gallery (WSG) mode and Fabry-Pérot (FP) mode respectively in NW, according to Yang *et al.* [17] If we compare Fig. 3 in this work with [17, Fig. 4], we can clearly observe that none of WSG, FP, or hybrid modes exist in the wavelength range which we are studying here. Moreover, a well designed nanowire can enhance the absorption of solar cells by incorporating WSG/FP resonances [18]. WSG mode lasing can be also achieved when adorning horizontal NWs with metal particles [19].

To gain further insight of the optical properties inside the NWs, we also studied the Purcell factors (F) of the PRK NWs with different diameters. When the media is lossless, F can be simply enumerated by the ratio of radiated power in the far zone (P) to the radiated power in the free space (P_0), namely $F = P/P_0$ [20]–[22] Lumerical FDTD can directly calculate the Purcell factors of the dipole source when the media is lossless dielectric. As we assume the real refractive index (imaginary part $k = 0$) at the emission wavelength 530 nm, the PRK NWs can be regarded as lossless. If the k is not 0, an extra transmission box surrounding the dipole source needs to be appended.

We calculate the Purcell factor of the horizontal PRK NWs with different wavelengths from 500 nm to 1,100 nm and assume the NW has a constant index of $n = 2.2$ and $k = 0$. Wavelength shorter than 500 nm is not included because PRK NW is absorptive below 500 nm. In the above-mentioned range, wavelengths of 530 nm and 1,060 nm are of extreme significance because they correspond to the emission wavelength and the largest excitation wavelength

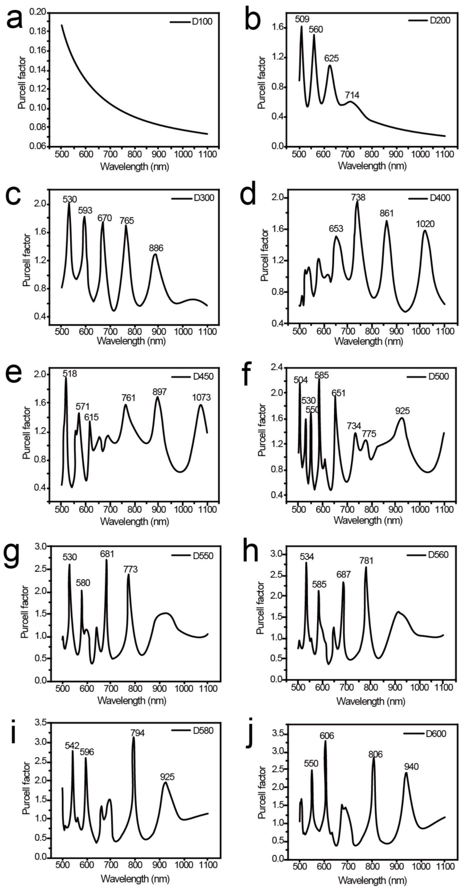

FIGURE 4. Purcell factors of the horizontal NWs with different diameters, TE mode. D means the diameter of the NW and the unit is nm.

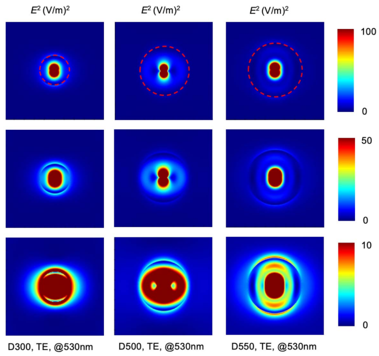

FIGURE 5. Radial-plane cross-sectional E^2 profiles of the NWs with diameters of 300 nm, 500 nm, and 550 nm (@530 nm with TE mode dipole).

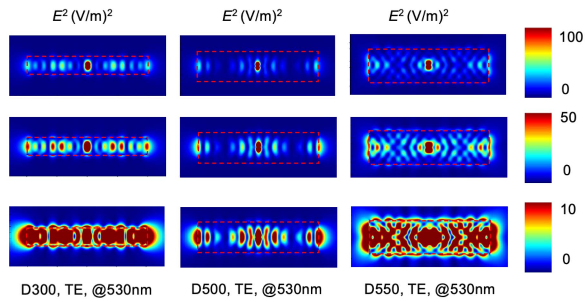

FIGURE 6. Axial-plane cross-sectional E^2 profiles of the NWs with diameters of 300 nm, 500 nm, and 550 nm (@530 nm with TE mode dipole).

for two-photon up-conversion respectively. In fact, photons with a wavelength smaller than 1,060 nm can excite the two-photon absorption [23], [24]. For instance, Waters *et al.* pumped the $CsPbBr_3$ single crystals with 800 nm light and generated 570 nm emission [25]. Also up-conversion has a lot important applications such as two photon lasing [26]–[28].

Fig. 4 shows the F of the horizontal PRK NWs with TE mode dipole source. For NW with 100 nm diameter, there is no resonance peak, which means no resonance mode is supported in PRK NW with such a small diameter. When the NW diameter increases, more resonance peaks start to emerge. It's worth noting that NWs with diameters of 300 nm, 500 nm, and 550 nm all have prominent resonance peaks at the wavelength of 530 nm, which confirms the potency of the NWs to be used as a lasing cavity [29], [30]. NW with a diameter of 400 nm has a resonance peak near 1,020 nm, a little less than 1,060 nm, rendering the NW a candidate for a two-photon up-conversion study.

The previous absorption spectra do not show strong resonance. The reason might be the difference in the light source. The absorption is simulated with plane wave incident onto the NW and the F is simulated with the dipole source located inside NW.

In order to get a better understanding of the resonance peak at 530 nm wavelength in the F spectra, we simulate the radial-plane cross-sectional E^2 profiles at the center of the NWs with diameters of 300 nm, 500 nm, and 550 nm, as shown in Fig. 5. Three different color bar ranges of 0-100 $(V/m)^2$, 0-50 $(V/m)^2$, and 0-10 $(V/m)^2$ are shown from the top row to the bottom row. Distinct E field enhancement at the NW boundaries can be resolved.

Moreover, we also simulate the axial plane cross-sectional E^2 profiles, as shown in Fig. 6. E field enhancement at the end facets of NWs can be resolved, which indicates the resonance at 530 nm wavelength in the F spectra corresponds to the leaky modes. The NWs with diameters of 300 nm, 500 nm, and 550 nm work as antennas at the wavelength of 530 nm.

It's worth noting that the resonance peaks are sensitive to the index of the PRK NW. If the NW is made of mixed PRK and PEO, the index can be in the range of 1.5-2.2, as mentioned in the previous discussion. The above discussion is based on pure PRK. For mixed PRK/PEO, the resonance peaks would shift and require different optimal geometries.

Moreover, the F of the NW with TM mode dipole source is shown in Fig. 7. For the TM mode dipole source, the electrical field polarization is along the axial direction of the NW. It is obvious that no distinct resonance is observed for NWs with diameters below 300 nm. And only one resonance peak exists at 553 nm for the NW with a diameter of 300 nm.

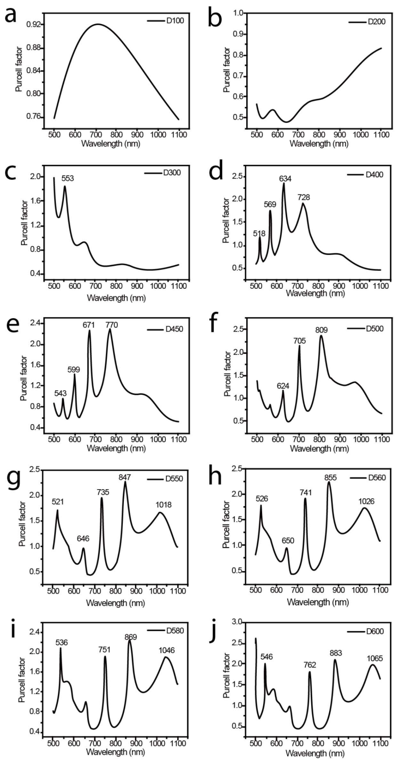

FIGURE 7. Purcell factors of the horizontal NW with different diameters, TM mode dipole source. D means the diameter of the NW and the unit is nm.

Even though there are multiple resonance peaks in NWs with the diameters larger than 300 nm, no resonance peak appears at the wavelength of exact 530 nm. For NWs with diameters of 550 nm, 560 nm, and 580 nm, resonance peaks near and smaller than 1,060 nm can be observed, rendering those diameters suitable for two-photon absorption. According to Zhou *et al.*, TM mode dipole can excite FP modes in NWs, which in turn can enhance light emission [31]. This can explain why LEE_{TM} is larger than LEE_{TE}.

Besides, we propose the following fabrication processes for the horizontal PRK NW LED devices. First, utilizing shadow masks, Ag/LiF lines can be thermally evaporated onto the patterned ITO glasses (commercially available). Second, lines of PEDOT:PSS and TPBi can be deposited by ink-jet printing and thermal evaporation (with shadow masks) methods onto ITO and Ag respectively. Then the synthesized PRK NWs precursor solution can be deposited onto the substrates to adhere to the injection layers either by drop-casting or ink-jet printing. Polymers such as PEO or polyimide can be mixed with the PRK NWs precursor solutions to further engender the stability of PRK NWs and meanwhile reduce the refractive index of the active material.

Limited by our current fabrication processes, we have not achieved the real horizontally aligned nanowire-based LEDs with high performance but hopefully we can make it in the near future.

Regarding the amount of emanating light, there are a few points which need to be lucidly told. If the injected current is the same for horizontal NW and planar LEDs, the injected carriers are the same. Then the amount of emanated light depends on the EQEs of the two structured LEDs. Moreover, if the two structured LEDs work at the same driving voltage, the planar one will have a much larger injected current considering the horizontal NW theoretically has much higher resistance.

$$R_{horizontal\ NW} = \rho \frac{L}{\pi r^2} \tag{3}$$

$$R_{planar} = \rho \frac{t}{A} \tag{4}$$

where ρ is the electrical resistivity, L is the NW length, r is the NW radius, t is the planar perovskite film thickness, and A is the perovskite film area. Here, if we use $L = 2\ \mu m$, $r = 225\ nm$, $t = 50\ nm$, and $A = 2\ mm^2$, (225 nm is the optimal radius from Table 1, and 50 nm thickness and 2 mm^2 are the typical perovskite thickness and device area for planar LEDs respectively.) we can find that

$$R_{planar} \approx 2 \times 10^{-9} \cdot R_{horizontal\ NW}.$$

The comparison will be more just if we use 2 μm thickness (equal to NW diameter) and $\pi \cdot (225\ nm)^2$ device area for planar structure but the thickness is far from the optimal value and the device area is too small to be realized practically.

A much larger injected current will lead to more injected carriers and more extracted photons in the planar device. The situation can be different if we have parallel horizontally aligned NW arrays, but it's beyond the scope of this work.

III. CONCLUSION

In this work, we propose a horizontally aligned PRK NW LED device with a high LEE of 50.9%, and a 2.9-fold enhancement compared to the planar counterpart. The NW geometry effect has also been systematically studied. Both absorptions of the plane wave and emission of the dipole source are studied with TE and TM modes. For plane-wave absorption, there are no strong absorption resonances in the range of 400-600 nm. For dipole emission, resonance peaks in the F spectra exist at 530 nm wavelength with TE mode dipole for NWs with diameters of 300 nm, 500 nm, and 550 nm respectively. After studying the E^2 profiles, we deciphered that the resonance peak is connected with the antennas effect. We also propose the fabrication processes of horizontally aligned PRK NWs based on the ink-jet printing method. This work paves the way for a novel design of the horizontally aligned PRK NW LED devices, and the associated light extraction mechanisms are also beneficial to the future design of other light emission devices such as lasers.

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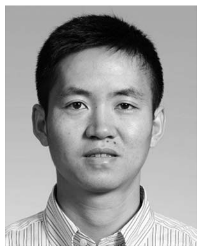

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