

Microcavity Design Upping Light Extraction Efficiency over 50% in High-Index Perovskite Light-Emitting Diodes

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Perovskite light-emitting diodes (PeLEDs) are promising candidates for lighting and display applications. However, perovskites usually have a relatively high refractive index compared to organic semiconductors, causing a lower optical efficiency due to a narrower escape cone. In this work, the theoretical analysis shows that the microcavity effect enables PeLEDs to achieve a light extraction efficiency of 51%, much higher than the $\approx 20\%$ estimated by the classical theory. Besides the interference in the microcavity, the efficiency improvement is also attributed to the low surface plasmon loss and a high ratio of horizontal dipoles under the Purcell effect. In a microcavity PeLED, a horizontal dipole shows three times the improvement in the Purcell factor of a vertical dipole, making the horizontal dipole with high efficiency contribute 86% of light output. Moreover, the experiment shows that the microcavity can increase the external quantum efficiency by 70%. A narrower spectrum and shorter photoluminescence lifetime are also observed. These phenomena are attributed to the simultaneous enhancement of light extraction efficiency and internal quantum efficiency. This work reveals the importance of microcavity design and provides guidance to break through the classical efficiency limit and achieve high-efficiency PeLEDs.

1. Introduction

Metal halide perovskite materials exhibit superior optoelectronic properties, such as narrow spectral width, wide-range tunable bandgap, and high luminescence quantum yield. Compared to organic light-emitting diodes (OLEDs), perovskite light-emitting diodes (PeLEDs) show higher color purity and widely tunable bandgap,^[1,2] making them promising candidates in lighting and display applications. Over the past few years, PeLEDs have been rapidly developing and have achieved an external quantum efficiency (EQE) beyond 20%.^[2–5] In contrast, the planar OLEDs have performed with an EQE close to 30%^[6–11] after more than 30 years of development.^[12] For the practical application of PeLEDs, an EQE comparable to that of OLEDs is desirable. The material engineering and fabrication process of PeLEDs have been extensively studied to achieve high efficiency, while optical factors have only gained attention in recent years.

To unlock the full potential of PeLEDs, this work will investigate microcavity design and discuss the optical efficiency limit. The EQE of PeLEDs is given by^[13–15]

$$\eta_{\text{EQE}} = \eta_{\text{IQE}} \times \eta_{\text{LEE}} \\ = \eta_{\text{inj}} \times \eta_{\text{PLQE}} \times \eta_{\text{LEE}} \quad (1)$$

where η_{IQE} is the internal quantum efficiency (IQE), η_{inj} is the carrier injection efficiency, η_{PLQE} is the photoluminescence quantum efficiency (PLQE), and η_{LEE} is the light extraction efficiency (LEE) defined by the ratio of the photons emitted to free space to all photons generated at the active region. The high η_{inj} and η_{PLQE} can be realized by balancing and increasing charge injection and minimizing nonradiative recombination, respectively. Many efforts have been devoted to charge balance;^[16–19] the PLQE near 100% for perovskites was also reported.^[20–22] This indicates that a nearly 100% IQE can be potentially realized in PeLEDs through material and carrier injection structure engineering.^[23] However, LEE is severely restricted by total internal reflection (TIR), which is determined by device configuration. A general schematic PeLED architecture is shown in **Figure 1a**, where most light is trapped in the device by TIR because the refractive index of the

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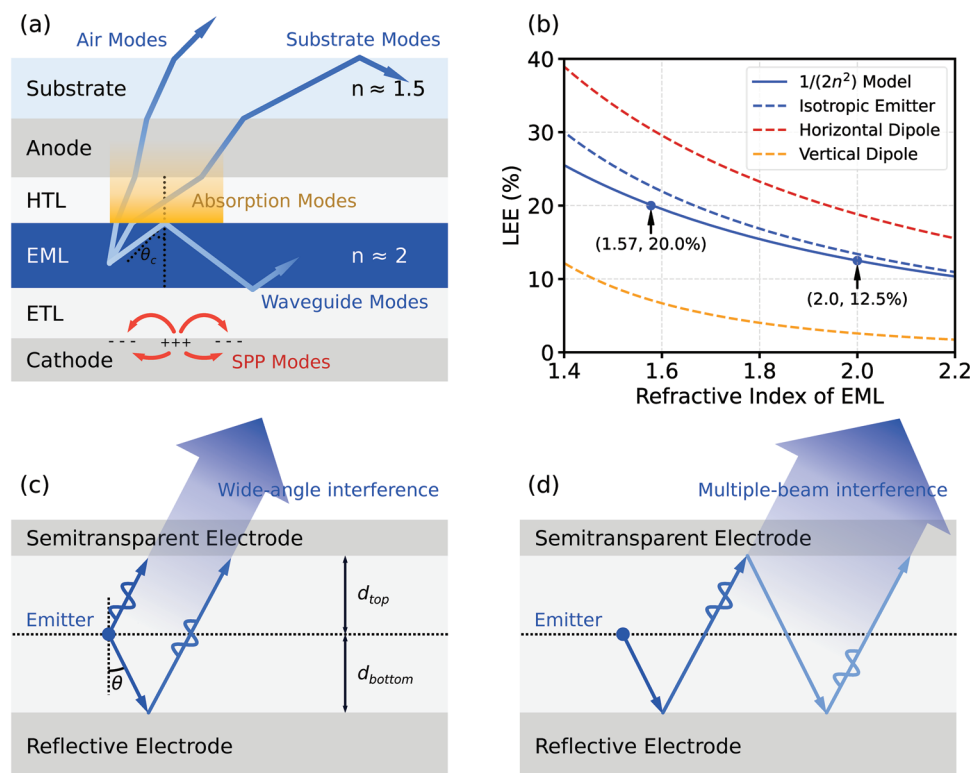


Figure 1. a) A typical device architecture of bottom-emission PeLEDs and its optical power distribution. b) $1/(2n^2)$ model and LEE approximation for oriented dipoles. c) Wide-angle and d) multiple-beam interference in a microcavity.

light emission layer (EML) is higher than air. For an isotropic emitter, an approximation^[24,25] of LEE is given by the ray-optics limit of $1/(2n^2)$. The corresponding relation is plotted as the blue solid curve in Figure 1b, which shows the LEE decreases as the refractive index of EML (EML index) increases. Generally, for planar LEDs without light extraction structures, some studies mentioned that the efficiency limit of OLEDs is about 20%^[26,27] because organic semiconductor materials usually have a refractive index of 1.6–1.8^[28] in the visible spectrum. Accordingly, it is reported that a low-index host layer can improve LEE.^[29,30] Unfortunately, perovskites show a higher index (2.0–3.2) than organic semiconductors, especially when they exist in bulk form.^[31–36] The result is shown in Figure 1b, the LEE limit of PeLEDs should be lower than 12.5% according to the $1/(2n^2)$ limit.

It is worth noting that the $1/(2n^2)$ model is based on the classical ray optics, and assumes that the device has a Lambertian emission pattern.^[24] It is reliable in the absence of interference. However, the nanoscale thickness of PeLED stacks tends to introduce interference and near-field effects. There are two significant effects, the microcavity effect and the Purcell effect, that also influence efficiency. The microcavity effect refers to the enhancement or inhibition of radiance due to constructive or destructive interference.^[37] As shown in Figure 1c,d, the microcavity introduces wide-angle and multiple-beam interference.^[38–40] The former is the interference between directly emitted and reflected waves with the same wave vector, while the latter describes that the light is reflected back and forth and superposed with itself. The Purcell effect describes the enhancement or inhibition of the spontaneous decay rate of an emitter in the device struc-

ture compared to that in free space.^[41,42] Based on the Purcell effect, we expect enhanced spontaneous emission in microcavity structures.

These effects have been discussed in OLEDs,^[43–48] while only few studies^[14,49] on microcavity PeLEDs are reported. One of the studies reported that the 3D and quasi-2D PeLEDs can achieve maximum theoretical EQEs of 25% and 20%, respectively, in spite of the high EML index.^[50] However, these effects in PeLEDs have rarely been systematically analyzed. In this context, we propose that microcavity design in PeLEDs could improve LEE to over 40% and even close to 50%. Three factors contribute to such high efficiency in PeLEDs. The first is the angular-dependent intensity of microcavity; we can concentrate emission within the escape cone. The second is the high ratio of horizontal dipoles when the Purcell factor of the horizontal dipole is larger than the vertical dipole. The non-uniform Purcell factors indicate that the microcavity PeLED exhibits a selective emission enhancement in different dipole orientations. Our results show that the efficiency of an isotropic emitter in the microcavity can be only 15% less than that of an anisotropic emitter consisting of only horizontal dipoles. It indicates that the microcavity is an alternative approach to produce anisotropic emission without orienting and aligning anisotropic nanocrystals, such as rods and platelets. By contrast, in OLEDs, controlling molecular orientation is a general strategy for improving efficiency.^[51–53] The third factor is the less surface plasmon polaritons (SPPs) loss due to the high EML index of PeLEDs, indicating that more light is under the manipulation of microcavity. Interestingly, in this case, LEE could increase as the EML index increases, which is the opposite of the

$1/(2n^2)$ model. This work reveals that microcavity design makes it possible for PeLEDs to outperform OLEDs in efficiency.

2. Microcavity Optimization with Semitransparent Electrode

A typical PeLED structure shown in Figure 1a is composed of a reflective cathode, an electron transport layer (ETL), an EML, a hole transport layer (HTL), an anode, and a substrate. Because of the high refractive index difference among layers and the limited thickness, PeLEDs naturally construct a microcavity. For simplicity, we combine the dielectric stack into one layer in Figure 1c,d, in which the microcavity resonance condition is given by

$$\frac{2\pi}{\lambda} 2nd_{\text{bottom}} \cos \theta - \phi_{\text{bottom}} = m2\pi \quad (2)$$

$$\frac{2\pi}{\lambda} 2nd_{\text{top}} \cos \theta - \phi_{\text{top}} = m2\pi \quad (3)$$

$$\frac{2\pi}{\lambda} 2n(d_{\text{bottom}} + d_{\text{top}}) \cos \theta - (\phi_{\text{bottom}} + \phi_{\text{top}}) = m2\pi \quad (4)$$

where λ is the wavelength of emission in a vacuum, n is the averaged refractive index of layers in device structure, d_{top} and d_{bottom} are the distances between the emitter to the corresponding reflective interface, θ is the angle of propagation, ϕ_{top} and ϕ_{bottom} are the phase shifts at the top and bottom interface, respectively, and $m = 0, 1, 2, \dots$ is the resonance order. Equations (2) and (3) describe that the wide-angle interference has a strong dependence on d_{bottom} and d_{top} , that is, the position of the emitter. Equation (4) shows that the multiple-beam interference is related to the total cavity length $d_{\text{bottom}} + d_{\text{top}}$. In bottom-emission PeLEDs, the semitransparent electrode is usually a conductive oxide with high transparency, such as indium tin oxide (ITO). Therefore, the reflection from the semitransparent electrode is very weak in the escape cone, and the effects of Equations (3) and (4) are limited. In other words, the wide-angle interference described by Equation (2) is dominant in common bottom-emission PeLEDs,^[38,39] and we can call those PeLEDs the weak-microcavity PeLED.^[54,55] On the contrary, a strong-microcavity PeLED can be formed if the semitransparent electrode is a thin metal film, such as silver (Ag) or aluminum (Al). As a result, the multiple-beam interference associated with Equation (4) can be enhanced and manipulated in the microcavity. A similar structure can also be used for top-emission PeLEDs. Since the light extraction competes with optical losses, we should optimize the microcavity to ensure constructive interference within the escape cone while destructive interference occurs in other directions.

Considering the multilayer structure of PeLEDs, the oscillating dipole model based on the Chance, Prock, and Sibley (CPS) theory^[56] is applied to analyze the optical properties of PeLEDs. The studied PeLED is with the structure of glass / ITO / semitransparent electrode (optional) / MoO_3 (3 nm) / PEDOT:PSS (30 nm) / MoO_3 (3 nm) / PolyTPD (20 nm) / perovskite (30 nm) / TPBi (35 nm) / LiF (1 nm) / reflective electrode (100 nm), and the peak emission wavelength is 690 nm. To enhance the microcavity effect, we insert a thin metal film as the semitransparent electrode between ITO and MoO_3 . In this case, we can compare the structure to a Fabry–Perot optical resonator without losses. At

Table 1. LEE optimization of the microcavity PeLEDs with Al and Ag as the electrodes, respectively, at wavelength of 690 nm. d represents thickness; the legend of the power distribution is consistent with Figure 2b.

Layer	Min. d [nm]	Max. d [nm]	Optimal d [nm]	Power distribution
Al as the semitransparent and reflective electrodes				LEE _{max} = 30.7%
Thin Al film	1	30	2.8	
PEDOT:PSS	10	80	44.6	
TPBi	10	80	48.5	
Ag as the semitransparent and reflective electrodes				LEE _{max} = 51.4%
Thin Ag film	1	30	22.0	
PEDOT:PSS	10	80	21.4	
TPBi	10	80	53.7	

the resonance frequency, the emission enhancement factor G_e in the normal direction is given by^[57–59]

$$G_e \propto \frac{(1 + \sqrt{R_2})^2 (1 - R_1)}{(1 - \sqrt{R_1 R_2})^2} \quad (5)$$

where R_1 and R_2 are the reflectances of semitransparent and reflective metal electrodes, respectively. This equation indicates that the reflectance of the reflective electrode should be as large as possible for emission enhancement. Figure 2a shows the calculated reflectance of 100 nm-thick Al, Ag, and Au films at normal incidence. We can find that Ag offers the highest reflectance at 690 nm, so it is a good reflective electrode for our microcavity PeLED. We also choose Ag as the semitransparent electrode and optimize its thickness d to improve efficiency. According to Equations (2)–(4), the cavity length and the emitter's position are vital for a microcavity; here, we choose PEDOT:PSS and TPBi thickness as variables, and their range is limited from 10 to 80 nm to support charge injection. We assume the refractive index of perovskite film is 2.4 as an appropriate median.

An optimization algorithm based on quadratic approximation is employed to find the maximum LEE at the wavelength of 690 nm (red light), and the results with different electrodes are shown in Table 1. First, we can observe that the maximum LEE reaches 51.4% in the microcavity PeLED with Ag as the semitransparent and reflective electrodes; this value is 1.67 times the optimal efficiency with Al electrodes. Second, the optimal thickness of thin Al film is as low as 2.8 nm, implying that the benefit of the strong microcavity with Al is limited, and the efficiency does not exceed that of the weak microcavity. The reason for such a gap is the reflectance difference of electrodes. Even though the reflectance difference between Al and Ag films is only 7.9% (Figure 2a), the losses are amplified during circulation, and the enhancement would decrease dramatically. Thus, choosing appropriate electrodes is essential when designing microcavity PeLEDs. Overall, our results show that the 51.4% efficiency under the microcavity is almost six times the 8.7% efficiency estimated by the $1/(2n^2)$ model when the EML index is 2.4.

To further illustrate the principle of the microcavity, the thickness of semitransparent Ag is swept in the optimized PeLED with 21.4 nm PEDOT:PSS and 53.7 nm TPBi. When the

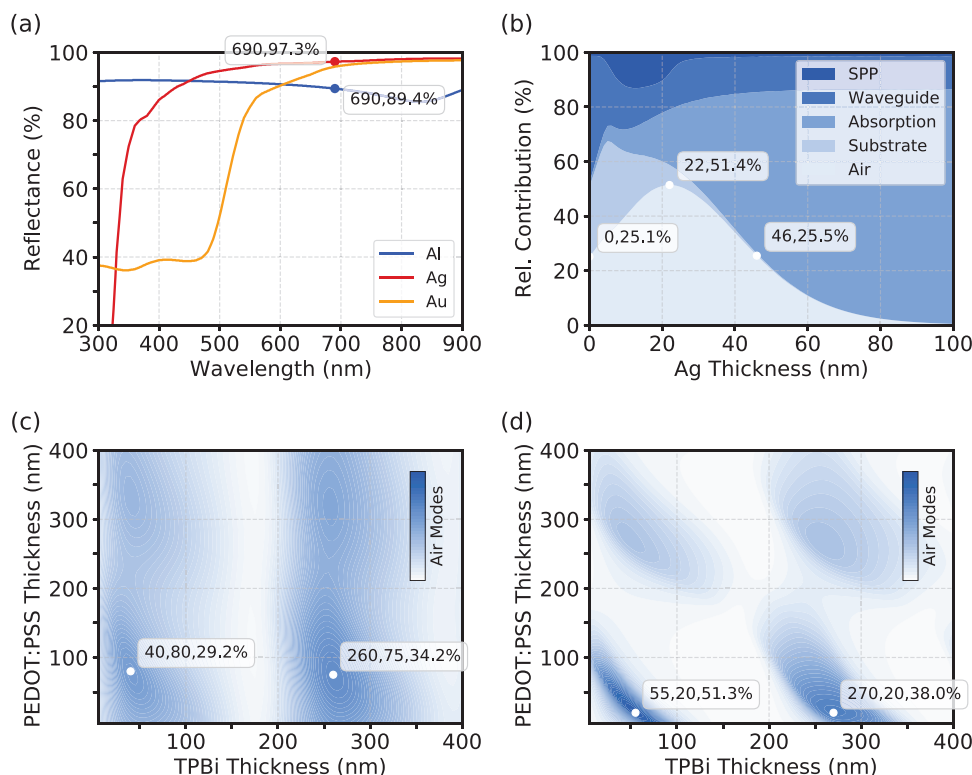


Figure 2. a) Calculated reflectance of 100 nm thick Al, Ag, and Au at normal incidence. b) Power distribution of different optical modes versus Ag thickness. Air modes of c) the weak-microcavity PeLED and d) the strong-microcavity PeLED versus TPBi and PEDOT:PSS thickness variation.

Ag thickness increases, its transmittance decreases; more light is supposed to be trapped in the device. However, Figure 2b shows that when the Ag thickness increases from 0 to 22 nm, the efficiency increases dramatically until it has been doubled (for details of optical modes, see Figure 1a and Section S1, Supporting Information). This phenomenon is attributed to the constructive interference within the escape cone when the microcavity resonance condition is carefully adjusted. Correspondingly, the waveguide modes outside the escape cone are limited due to destructive interference. When the thickness of Ag exceeds 22 nm, the efficiency starts to decrease because the Ag with decreased transmittance traps more and more light, and the absorption modes continue to rise. When the thickness of Ag reaches 46 nm, the enhancement of microcavity is offset by the absorption, and the efficiency returns to the initial value.

The microcavity resonance can be modified by the thickness of ETLs and HTLs. In the weak-microcavity PeLED with Ag as the reflective electrode, the dependence of air modes on the thickness of TPBi and PEDOT:PSS is plotted in Figure 2c. As we discussed, the wide-angle interference described by Equation (2) is dominant in the weak-microcavity PeLED. Therefore, Figure 2c shows the high variability of efficiency by changing TPBi thickness, while the PEDOT:PSS thickness variation hardly affects the efficiency. The maximum efficiency of 34.2% occurs in the second resonance order, because the angular intensity distribution is wide in the first resonance order, and the substrate modes share the constructive interference. For the second order, however, it is challenging to achieve 260 nm thick TPBi technically while maintaining good electrical conductance. By contrast,

in the strong-microcavity PeLED shown in Figure 2d, maximum efficiency of 51.3% can be obtained in the first order with good feasibility, which is a good tradeoff of LEE and carrier transport. In this process, efficiency fluctuates more dramatically, and the PEDOT:PSS thickness also strongly influences the efficiency owing to the enhanced multiple-beam interference. Therefore, a higher requirement for precise thickness control is needed to achieve the target efficiency.^[60] Electrically, the 55 nm TPBi may have some adverse effects on carrier transport. We can appropriately reduce the TPBi thickness and increase the PEDOT:PSS thickness to achieve a similar cavity length. As for green and blue emissions, the optimal ETL thickness would be smaller because of the shorter cavity length required for resonance.

3. Selective Enhancement of Dipole Emission

We have discussed the interferences in a microcavity, which depend on the position of the emitter and the cavity length. Interestingly, the decay rate is also position-dependent in a microcavity. Because the field generated by the dipole would be reflected by interfaces, especially the metallic interface, then the reflected field becomes a driving force to the dipole.^[61] The dynamics of the dipole is given in Section S2, Supporting Information. If the reflected field is in phase with the dipole, the dipole oscillates harder with a higher radiative decay rate b . If the reflected field is out of phase, b decreases, and the radiative decay is inhibited. The phase is distance-dependent, which is similar to the interference in the microcavity. Therefore, the radiative decay can also be modified by the microcavity. The modification of the radiative

decay rate is the Purcell factor F , which is given by^[45]

$$F = \frac{b}{b_0} \quad (6)$$

where b_0 is the radiative decay rate in the absence of any reflection.

If the intrinsic quantum efficiency of the dipole q_0 is not unity, there is competition between radiative and nonradiative recombination. An enhanced radiative decay can improve the quantum efficiency. Thus, the quantum efficiency is also modified by the Purcell factor^[45,62]

$$q_{\text{eff}} = \frac{b}{b + b_{\text{nr}}} = \frac{b_0 F}{b_0 F + b_{\text{nr}}} = \frac{q_0 F}{q_0 F + 1 - q_0} \quad (7)$$

where q_{eff} is the effective quantum efficiency, b_{nr} is the nonradiative decay rate. If F is greater than 1, q_{eff} is larger than q_0 . This implies that an optimized microcavity can even improve IQE in addition to improving LEE.

In order to investigate the efficiency limit, IQE is assumed to be 100% in our simulation. However, it is still valuable to investigate the role of microcavity in modifying the radiative decay rate. We will reveal that the microcavity has the ability to modify the ratio between horizontal dipoles and vertical dipoles, which facilitates extremely high efficiency. In general, a horizontal dipole has a higher LEE than a vertical dipole because its emission is more concentrated in vertical direction. The emission intensity of a dipole relates to $\sin^2(\theta)$ (see Section S3, Supporting Information), where θ is the angle between the dipole axis and the direction of light emission. The intensity versus θ is normalized and shown in Figure 3a. We use the transverse magnetic (TM) plane to give the 2D cross-section of dipole radiation patterns. It can be observed that the horizontal dipole emits more photons within the escape cone and has a higher LEE than the vertical dipole. For the isotropic emitter, nearly 80% of light is in waveguide modes. However, in the strong-microcavity PeLED, the angular dependence of emission is strong, as plotted in Figure 3b, most of the light can be concentrated within the escape angle. It should be noted that the maximum intensity for optimal efficiency is not in the normal direction because the solid angle ($d\Omega = \sin\theta d\theta d\varphi$) is small near the normal direction ($\theta \approx 0$).^[39] It is worth noting that although non-Lambertian emission may not be suitable for planar display, it can be potentially applied to augmented reality (AR) and virtual reality (VR) displays owing to the concentrated emission and the high efficiency.

In addition to concentrating light inside the escape cone, microcavity also plays a role in regulating the decay rate of dipoles with different orientations. Figure 1b shows that the low LEE of the vertical dipole is an obstacle for the total LEE (see Section S4, Supporting Information). If the vertical dipoles are inhibited, while the horizontal dipoles are enhanced, the overall LEE will approach the high LEE of the horizontal dipole. The LEE of an isotropic emitter is related to the Purcell factor of the vertical and horizontal dipoles, which is given by

$$\text{LEE} = \frac{P_{\text{ext}}}{P_{\text{dis}}} = \frac{F_v \text{LEE}_v + 2F_h \text{LEE}_h}{F_v + 2F_h} \approx \frac{2F_h}{F_v + 2F_h} \text{LEE}_h \quad (8)$$

where P_{dis} and P_{ext} are the dissipated power and extracted power, respectively; F is the Purcell factor representing the normalized

dissipated power, the subscripts v and h denote the vertical and horizontal dipole, respectively. Since LEE_v is much smaller than LEE_h in an optimized microcavity, $F_v \text{LEE}_v$ can be eliminated and the equation would be straightforward, increasing F_h and decreasing F_v is a key for LEE to approach the high LEE_h . If we can demonstrate that F_h is higher than F_v in the microcavity PeLED, horizontal dipole has a higher contribution to the total emission in microcavity PeLEDs. It will interpret why the microcavity can achieve 51.4% LEE, despite the escape cone only occupying 9.1% of the hemisphere when the EML index is 2.4.

The strong-microcavity PeLED is optimized as stated in Table 1. Figure 3c–e shows the optical power distributions and power dissipation spectra of the horizontal dipole, the vertical dipole, and the isotropic emitter, respectively. The higher effective refractive index n_{eff} in power dissipation spectra relates to a larger propagating angle (see Section S5, Supporting Information). Comparing Figure 3c–e, we can observe three phenomena. First, the vertical dipole's intensity distribution is mainly at large angles, as shown in Figure 3a, leading to power distribution in high n_{eff} (Figure 3d). Since the light emitted by the vertical dipole is TM-polarized, it is easily coupled to SPPs. As a result, the optical channel of the vertical dipole is mainly waveguide modes (WVG) and SPP modes. Such dissipation encumbers the overall LEE. By contrast, the horizontal dipole emits light at a small angle and small n_{eff} , less light is coupled to the waveguide; and half the light is transverse electric (TE) polarized,^[63] which cannot support the SPP excitation. As shown in Figure 3c, the air modes of the horizontal dipole can be up to 60%. Second, although the air modes of the vertical dipole are almost zero, the air modes of the isotropic emitter (51%) are still close to that of the horizontal dipole (60%). As we discussed in Equation (8), the enhancement of horizontal dipole decay by the microcavity results in a larger contribution of horizontal dipole to LEE. As shown in Figure 3f, at the wavelength of 690 nm, the Purcell factor of the horizontal dipole (F_h) is three times that of the vertical dipole (F_v). We can also find the larger Purcell factor variation for horizontal dipole relative to vertical dipole. Since the electric field of the horizontal dipole has more energy distribution in the vertical direction, the reflected field at the dipole position is stronger relative to the vertical dipole. As a result, the decay of a horizontal dipole is more affected by the microcavity. Substituting the Purcell factor into Equation (8), we can obtain that 86% of light is emitted by the horizontal dipole. If only efficiency is concerned, the microcavity can be an alternative to orienting the perovskite nanorods and nanoplatelets, to obtain a high ratio of horizontal dipole.^[64,65] Third, the wavelength tolerance is relatively high for the strong-microcavity PeLED, and more than 40% of air modes can be maintained in the wavelength range of 650–730 nm. For blue and green emissions, the wavelength tolerance would be smaller. Compared with OLEDs, PeLEDs' spectra are much narrower and can even achieve a full width at half maximum (FWHM) below 20 nm.^[66,67] This is another reason microcavities are more suitable for PeLEDs than OLEDs.

4. Impact of SPP Modes on Efficiency

The limited distance between the emitter and metal electrodes usually increases loss by SPPs and limits microcavities' performance. Although OLEDs' efficiency is affected by SPP modes

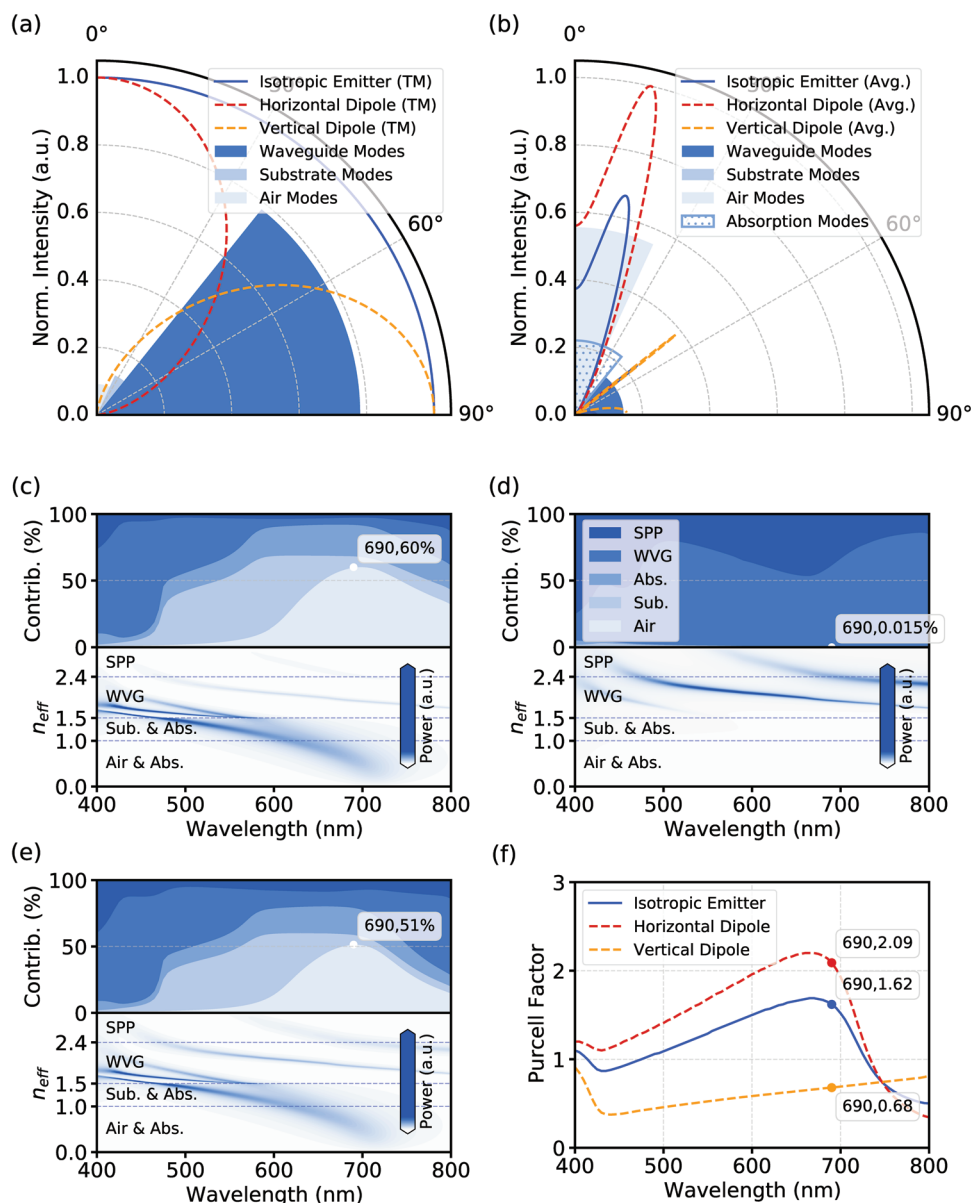


Figure 3. 2D cross-section views of the far-field dipole radiation patterns in a) free space and b) the microcavity PeLED. Optical power distributions and power dissipation spectra of c) the horizontal dipole, d) the vertical dipole, and e) the isotropic emitter in the microcavity PeLED. f) The Purcell factors versus wavelength for the strong-microcavity PeLED.

trapping up to 50% of the total power,^[54,68,69] SPP modes are limited in the PeLED (Figure 2b), leaving more room for manipulation in the microcavity. The detailed understanding can be explained by that the higher EML index allows PeLEDs to attenuate near field coupling to SPP modes.^[70,71] To explain this phenomenon, we need to focus on the near-field of dipole. Because the angular spectrum of a dipole is broad in the near field and it can provide extended wave vectors and couple with SPPs.^[72] As the distance between the dipole and metal decreases, energy is transferred to SPPs, and the lifetime of the dipole drops quickly. In other words, the near-field coupling can also modify the decay rate because of the fluorescence quenching near a metallic surface.^[71,73]

Although we can attenuate the near-field coupling by increasing the thicknesses of the HTLs and ETLs, the thickness is usually not arbitrarily adjustable for the concern of carrier transport. In this section, we demonstrate that the refractive index of EML can also affect the near-field coupling (discussion in Section S6, Supporting Information). When the EML index increases in the weak-microcavity PeLED, as shown in Figure 4a, the SPP modes drop dramatically, but the air modes do not improve that much. As an appearance, the decreased SPP modes are mainly caused by the extension of the k_{in} range of the waveguide modes, SPPs are just classified as waveguide modes. Therefore, for the EML index variation, we think that both SPP and waveguide modes are resonances in the cavity and can be considered as a whole.

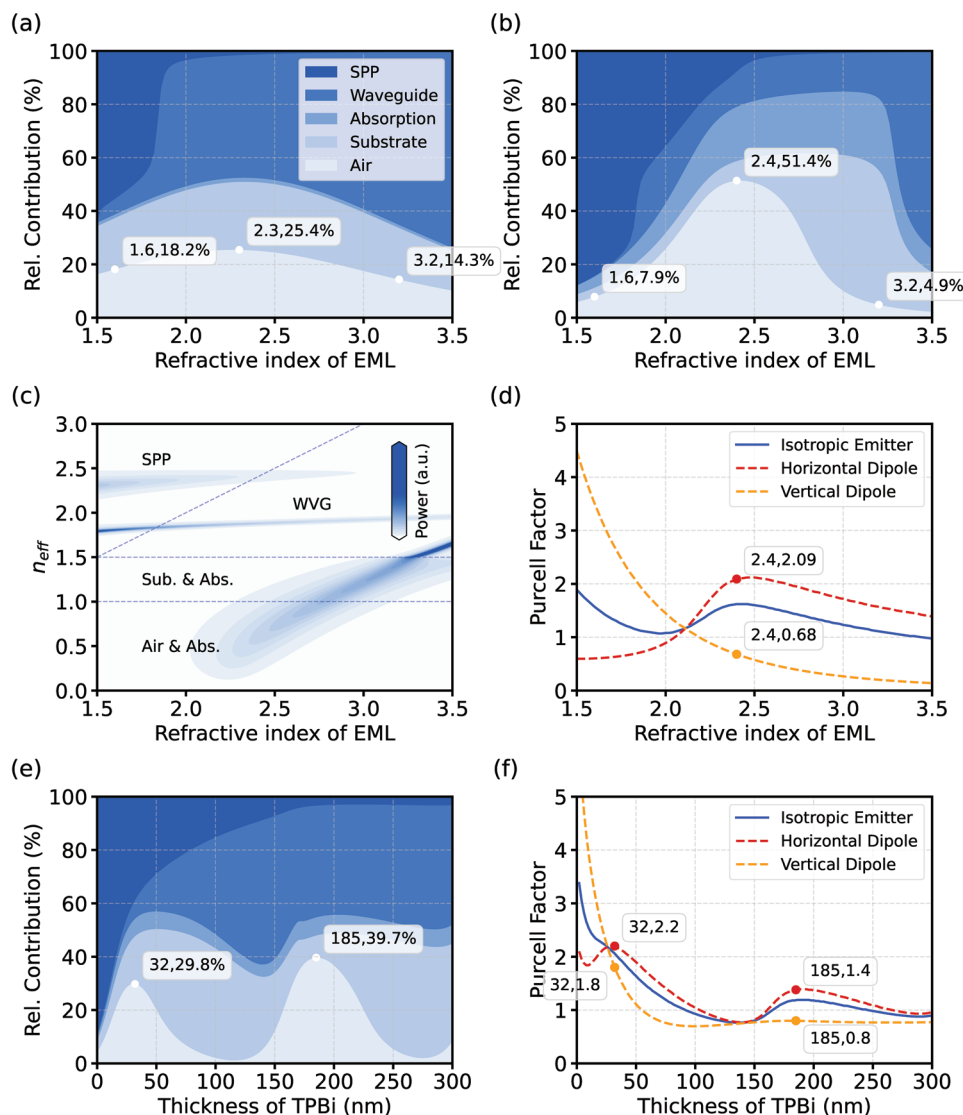


Figure 4. Optical power distributions versus the refractive index of the EML in a) the weak-microcavity PeLED and b) the strong-microcavity PeLED. c) Power dissipation as a function of EML for the strong-microcavity PeLED. d) The Purcell factor versus the refractive index of the EML in the strong-microcavity PeLED. e) Optical power distributions and f) the Purcell factor versus the TPBi thickness in the green NCs strong-microcavity PeLED.

Now we can observe that by increasing the EML index to 2.3 in Figure 4a, the air modes are slightly improved; we may attribute this to the weakened near-field coupling. If the EML index is larger than 2.3, the waveguide modes start to increase, and the air modes start to decrease because of the narrowed escape cone, which has been described by the $1/(2n^2)$ model. To clarify, we do not exclude the influence of the EML index on the resonance conditions; because the EML is very thin, the influence in the weak microcavity is limited.

As for the strong-microcavity PeLED, the effect of EML index is amplified as shown in Figure 4b. When the EML index increases from 1.6 to 2.4, both the SPP and waveguide modes decrease dramatically, while the air modes increase from 7.9% to 51.4%. This phenomenon completely contradicts the $1/(2n^2)$ model. However, if the EML index increases further, the $1/(2n^2)$ model starts to dominate, and the air modes will decrease. Unlike

the weak microcavity, the EML index has a significant influence on the resonance conditions in the strong-microcavity PeLED. Figure 4c presents the power dissipation versus EML index for the strong-microcavity PeLED. As the EML index increases, the emergence of the air modes is obvious. Meanwhile, the SPP is declining as the near-field coupling is attenuated. In Figure 4c, when the waveguide modes range covers SPPs as the EML index increases, the Otto and Otto–Kretschmann hybrid configuration for SPPs may be constructed in the far field. In this case, the surface plasmon resonance is damped because the waves satisfied SPPs are radiative in EML. Nevertheless, we think that the SPPs are mainly excited by the near field because the distance between the emitter and the metal is much smaller than the wavelength, and the near field is dominant. For the two reasons mentioned above, the increase in the air modes is not only related to the satisfied resonance conditions but also to the attenuated near-field

coupling. The Purcell factors are plotted in Figure 4d; the decay rate of the vertical dipole is high when the EML index is low; energy is dissipated to SPPs through the near-field coupling. The F_h and the F_v are equal until the EML index rises to 2.1. When the EML index increases to 2.4, the F_h achieves 2.09. Simultaneously, the F_v decreases to 0.68 due to the attenuated near-field coupling. In a word, the appropriate use of high-index EML in the microcavity not only reduces SPP losses but also increases the ratio of horizontal dipoles with high LEE.

The developed material engineering of perovskite provides us with index tunability. Except for bulk crystals or films, perovskites varied in morphology, such as nanoplatelets (NPLs), nanosheets (NSs), nanowires (NWs), and quantum dots (QDs).^[74] These perovskite NCs are usually capped with organic ligands for size control and surface defect passivation. The existence of ligands and spaces between NCs would reduce the refractive index of EML. In our experience, the index of NC film is usually 1.8–2.1, depending on the number and size of the ligands. We need to clarify that the optimal index of 2.4 is only appropriate for the above case. Individual optimizations are required for different device configurations and wavelengths, and sometimes the NCs with a lower index also perform well on LEE. For example, in blue or green NCs PeLEDs, there is a stronger SPP loss due to a shorter cavity length in the first-order microcavity. Figure 4e shows the green NCs PeLED with the same structure at the wavelength of 520 nm; the maximum efficiency occurs at a TPBi thickness of 185 nm, which is generally not feasible because the ETL and HTL thicknesses are limited for charge injection. In this case, we suggest adding a conductive spacer between the EML and the metal to relieve the near-field coupling. Accordingly, the spacer thickness also needs to be optimized to meet the microcavity resonance. Figure 4f shows the Purcell factor as a function of TPBi thickness; the F_v increases dramatically when TPBi thickness decreases due to near-field coupling. At the first local maximum efficiency, when TPBi thickness of 32 nm, the F_h is only 1.22 times the F_v . At the second local maximum efficiency, when TPBi thickness of 185 nm, the F_h is only 1.75 times the F_v . However, it is inevitable that the second-order microcavity will introduce more waveguide modes, so the efficiency cannot reach 50%.

Summarily, the EML index can influence efficiency through near-field coupling to SPPs and modification of the Purcell factor. These effects are deeply bound to the microcavity effect. A relatively high EML index can reduce the near-field coupling and enable the microcavity to achieve higher efficiency, which is contrary to the $1/(2n^2)$ model. From a traditional point of view, the high-index EML should be a fatal efficiency limitation for PeLEDs. Taking advantage of the microcavity, the weakness can be turned into an opportunity to exceed OLEDs in terms of efficiency. Consequently, a strong-microcavity PeLED can achieve a higher efficiency than OLED, in spite of the high EML index.

5. Experimental Verification of Microcavity Device

In order to verify the effectiveness of the microcavity, corresponding experiments are conducted. Since the standard of efficiency characterization is not consistent in some cases, we should specify the characterization method before presenting the results. First, our main measurement method is shown in Figure 5a. The emission surface is tightly attached to the port of the inte-

grating sphere. Consequently, only the photons that escape into the forward-viewing hemisphere are counted, while the photons from other sides are not.^[5,75,76] This measurement method is consistent with the definition of EQE, regardless of the $1/(2n^2)$ model or the dipole model. The second method is shown in Figure 5b; some studies may insert the whole device into the integrating sphere to characterize the EQE. This approach would collect photons from all surfaces; therefore, part of the substrate modes is also counted. The EQE obtained in this way may be useful for lighting but not for display applications. Obviously, it cannot correspond to either the theoretical model. The third method shown in Figure 5c utilizes a large-area detector and then puts the device very close to the detector with a small air gap.^[77] This approach complies with the EQE definition, but there will be an error from reflection at small angles or edge effects.^[78] Figure 5d shows the fourth method; it is usually used to measure the luminance at a certain angle. Suppose we use Lambertian approximation to obtain EQE according to the luminance at a single angle.^[3,79] In that case, it is obviously not applicable for microcavity devices because the emission of microcavity devices is strongly angle-dependent. Thus, we use the first two methods (Figure 5a,b) here to discuss the efficiency of microcavity devices. Refs. [77, 78, 80, 81] also provide some detailed discussions.

In the experiment, the EML is CsPbI₃ nanocrystals (NCs) film with a refractive index of 1.95 at wavelength of 690 nm (electroluminescence spectrum in Figure S1, Supporting Information). Two structures were fabricated to verify the effectiveness of the microcavity design. The weak-microcavity structure is glass / ITO / MoO₃ (3 nm) / PEDOT:PSS (40 nm) / MoO₃ (3 nm) / PolyTPD (20 nm) / CsPbI₃ NCs (30 nm) / TPBi (50 nm) / LiF (1 nm) / Ag (100 nm); the strong-microcavity structure is glass / ITO / Ag (10 nm) / MoO₃ (3 nm) / PEDOT:PSS (40 nm) / MoO₃ (3 nm) / PolyTPD (20 nm) / CsPbI₃ NCs (30 nm) / TPBi (50 nm) / LiF (1 nm) / Ag (100 nm). These two structures are optically and electrically optimized based on the practical photoluminescence spectrum. It ensures that both strong- and weak-microcavity structures are in a relatively good microcavity condition. The only difference between the two structures is the presence of a 10 nm Ag between ITO and MoO₃. We used two methods shown in Figure 5a,b to measure the EQE_{sur} from emission surface and EQE_{all} from all sides of the device.

As a result, the statistics of maximum EQE_{all} of the weak- and strong-microcavity PeLEDs are shown in Figure 5e. For comparison of optical efficiency, we selected a representative sample from each group. As shown in Figure 5f, the PeLED with weak microcavity (Ag group) shows the maximum EQE_{sur} of 8.51% at the applied voltage of 3.75 V, while the PeLED with strong microcavity (Ag–Ag group) shows EQE_{sur} of 14.42% at the applied voltage of 3.75 V. The presence of 10 nm Ag increases EQE by 70%. Another finding is that the ratio of EQE_{sur} to EQE_{all} of the Ag–Ag group is about 70%, which is higher than 51% of the Ag group. It indicates that more light is concentrated at small angles and emitted from the emission surface. If we obtain the intrinsic IQE (IQE_{int}) by fitting the simulated air modes and the experimental EQE_{sur}, the effective IQE (IQE_{eff}, i.e., the modified IQE under the Purcell effect) and substrate modes can be deduced. The theoretical and experimental results are shown in Table 2. First, both structures have a similar IQE_{int}, while air modes and EQE_{sur} of the strong microcavity far exceed the weak microcavity.

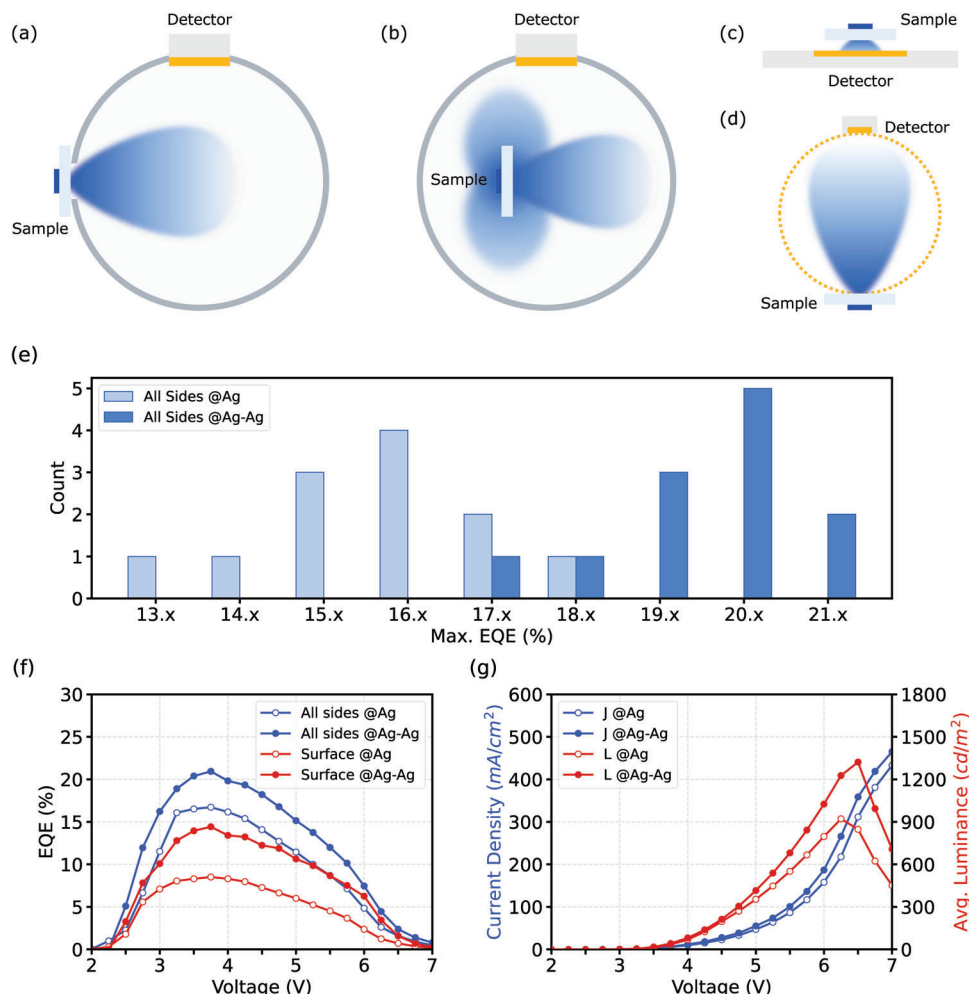


Figure 5. Experimental setups for efficiency characterizations. a) Integrating-sphere photometer measures photons emitted into the forward-viewing hemisphere. b) Integrating-sphere photometer measures photons emitted from all sides of a device. c) A large-area photometer close to a device measures photons emitted into the forward-viewing hemisphere. d) Luminance measurement at a certain angle. e) Statistics of maximum EQE_{all} measured by method (b) of the weak-microcavity (Ag) and strong-microcavity (Ag–Ag) PeLEDs. There are twelve samples for each group. f) Experimental EQE_{sur}–V characteristics and g) Experimental J–V–L characteristics of the weak- and strong-microcavity PeLEDs.

Table 2. Theoretical and experimental data of strong-microcavity and weak-microcavity PeLEDs. (Theoretical data: IQE_{int}, IQE_{eff}, air modes, Sub. + air modes; experimental data: EQE_{sur}, EQE_{all}).

IQE _{int}	IQE _{eff}	Air modes	EQE _{sur}	Sub. + Air modes	EQE _{all}
Ag as the reflective electrodes (weak microcavity)					
30.00%	34.31%	8.58%	8.51%	16.03%	16.72%
Ag as the semitransparent and reflective electrodes (strong microcavity)					
28.00%	37.72%	14.80%	14.42%	20.53%	20.93%

Second, although the intrinsic IQE is only $\approx 28\%$, the EQE_{sur} is up to 14.4% for the strong-microcavity PeLED. Third, the IQE of the strong-microcavity PeLED is enhanced by 34.7% from 28% to 37.72%; although its IQE_{int} of 28% is lower than that of the weak-microcavity PeLED, the situation for IQE_{eff} is reversed. The IQE enhancement can be explained by Equation (7). As a result, LEE as the ratio of EQE_{sur} (air modes) to IQE_{eff} is 25.0% and 39.2% for

the weak- and strong-microcavity PeLEDs, respectively; the LEE improvement is 56.8%.

As we discussed, EQE_{all} is not well defined and tends to be misused, but we are still interested in what it represents in our case. Interestingly, it can be observed that the sum of substrate modes and air modes is very close to EQE_{all} for both structures. Since the edges of the glass substrate are rough, we think that most of the substrate modes can leak out after scattering. In addition, because SPP modes are nonradiative and waveguide losses are very sensitive to absorption, only a minute portion of these modes can reach the edge of the device and emit light to free space. For these two reasons, we can think that the EQE_{all} of our devices measured by the method shown in Figure 5b is approximately the sum of substrate modes and air modes. The matching of EQE_{sur}–air modes and EQE_{all}–sub. + air modes can corroborate that the simulation matches the experiment and we think that the simulation accurately reflects the device's properties. It is worth noting that the rigorously defined EQE_{sur} is only about half of the EQE_{all} in the common PeLEDs with weak microcav-

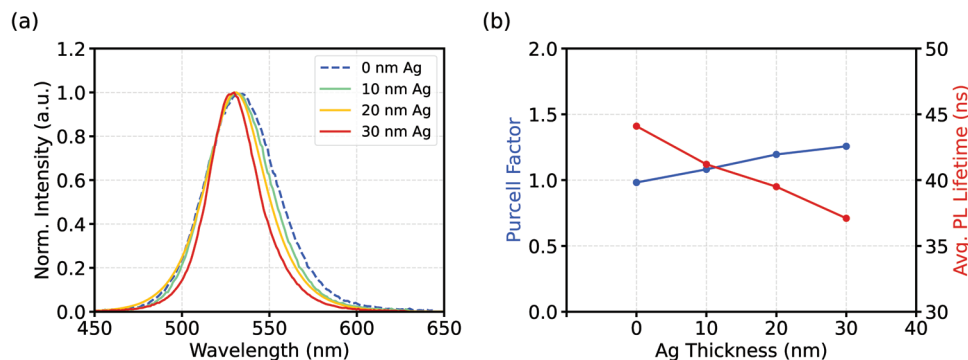


Figure 6. a) PL spectra. b) Purcell factor and average PL lifetime of PeLEDs with different Ag thicknesses.

ity. Thus, it is recommended to state the measurement method when the term EQE is mentioned. Figure 5g shows the current density–voltage–luminance curves of two structures; the maximum luminance is 1321.8 cd m^{-2} for strong-microcavity PeLED. Since we use the method shown in Figure 5a to measure the luminance, which is averaged over the hemisphere, the practical luminance in the emission direction should be higher for the strong-microcavity PeLED. Although the IQE of 28% is not competitive compared with state-of-the-art PeLEDs due to our immature materials and processes, the efficiency enhancement is sufficient to demonstrate the potential of the microcavity design in improving the optical efficiency of PeLEDs.

In addition, there is another experiment to verify the effect of Ag thickness on the spectrum and radiative decay rate. We fabricated PeLEDs with a structure of glass / ITO / semitransparent Ag (0, 10, 20, and 30 nm) / MoO_3 (3 nm) / PEDOT:PSS (20 nm) / MoO_3 (3 nm) / PolyTPD (20 nm) / bulk CsPbBr_3 (30 nm) / TPBi (55 nm) / LiF (1 nm) / Ag (100 nm), and the peak emission wavelength is about 530 nm. As shown in Figure 6a, the photoluminescence (PL) spectrum is narrowed as the thicker semitransparent Ag is applied. For the 0, 10, 20, and 30 nm Ag, the FWHMs of PL spectra are 47.8, 43.4, 39.5, and 32.8 nm, respectively. Based on the time-resolved photoluminescence (TRPL) characterization, we observed the shorter average PL lifetime as the Ag thickness increases as shown in Figure 6b. This phenomenon corresponds to the enhanced Purcell factor in the microcavity, according to the simulated blue line in Figure 6b. Combining Equations (6) and (7), the shorter PL lifetime implies enhanced quantum efficiency. Thus, we have experimentally demonstrated that microcavity has a significant impact on the light extraction efficiency, emission angle, spectrum, and quantum efficiency of PeLEDs.

Currently, a qualitative discussion of the experiment results is provided. To improve the quality of the work, it is better to present the theoretical results of the experimental devices together with the experimental results and provide an explanation. Finally, the conclusive statement is drawn. It's worth noting that Ag electrodes can push the upper limits of efficiency through microcavity optimization. However, improving EQE is a comprehensive project, influenced by factors such as material quality, electrical conditions, and optical conditions. Therefore, it is also important to address potential technical challenges that may arise from implementing Ag, such as energy band matching.

6. Conclusion

In conclusion, we systematically investigated the effect of microcavity on the efficiency of PeLEDs and demonstrated that an extremely high efficiency of over 50% is achievable. To explain the high efficiency, we demonstrated that the high ratio of the horizontal dipole with high efficiency is obtained in the strong-microcavity PeLED. Furthermore, we present that the efficiency can increase with increasing EML refractive index, which contradicts classical ray optics. The phenomenon is attributed to the attenuated near-field coupling and the selective enhancement of dipole emission. In the microcavity PeLED, the horizontal dipole shows three times the Purcell factor of the vertical dipole, indicating that the horizontal dipole emits 86% of light. We think that microcavity turns the disadvantage of high-index EML into an advantage and provides an opportunity for PeLEDs to surpass OLEDs in terms of efficiency. Accordingly, the experiment verified the optical performance improvement by microcavity optimization. We also observed a shorter average PL lifetime due to the enhanced Purcell factor in the microcavity PeLEDs. The experimental results show that the microcavity can improve LEE and IQE simultaneously.

7. Experimental Section

Device Fabrication: The ITO coated glass substrates were sequentially cleaned with detergent, deionized water, acetone, and ethanol by ultrasonic spin cleaning. The cleaned substrates were dried with N_2 flow and then treated by UV–ozone for 15 min. By using thermal evaporation under high vacuum ($\approx 1 \times 10^{-5} \text{ Pa}$), Ag (10 nm), and MoO_3 (3 nm) were deposited on the ITO at a rate of 0.05 and 0.002 nm s^{-1} , respectively. After the UV–ozone treatment, PEDOT:PSS (Clevios PVP AI4083) was spin-coated onto the ITO glass at 3500 rpm for 40 s followed by thermal annealing at 140°C for 20 min in the air. Another 3 nm MoO_3 layer was deposited on PEDOT:PSS at a rate of 0.002 nm s^{-1} . The hole transport layer of poly-TPD (4 mg mL^{-1} , in chlorobenzene) was spin-coated on top of the MoO_3 layer at 3000 rpm ($1500 \text{ rpm min}^{-1}$ accelerated speed) for 40 s in a N_2 filled glove box, followed by thermal annealing at 130°C for 30 min. Perovskite NCs (prepared according to ref. [82]) were dispersed in octane solvent and sequentially spin-coated onto the poly-TPD layer (2000 rpm for 40 s) to fully cover the underneath layer. Subsequently, TPBi (50 nm), LiF (1 nm), and Ag (100 nm) were deposited by thermal evaporation. The active area of the PeLED was 4.25 mm^2 , defined by the intersection of ITO and Ag strips.

Characterizations: Steady-state and TRPL were recorded with a PicoQuant FluoTime 300 spectrometer and a laser diode with a peak

wavelength of 510 nm was used as the excitation source. Thin films deposited on cleaned quartz substrates were used for these PL measurements. PLQYs were also determined from the thin films using the FluoroTime 300 spectrometer coupled with an integrating sphere. The PeLEDs were characterized in the N₂ filled glove box without encapsulation at room temperature. A Keithley 2635 source meter and a fiber optic integration sphere (Ocean Optics FOIS-1) coupled to a spectrometer (Ocean Optics QE65000) were used for the spectrum, luminance, and EQE measurements. The measurement system was calibrated by a standard tungsten halogen light source (Ocean Optics HL-3P-INT-CAL).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

external quantum efficiency, light extraction efficiency, light-emitting diodes, microcavity, perovskites

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