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Optical Energy Losses in Organic–Inorganic Hybrid Perovskite Light-Emitting Diodes

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Light-emitting diodes (LEDs) based on organic–inorganic hybrid perovskites, in particular, 3D and quasi-2D ones, are in the fast development and their external quantum efficiencies (EQEs) have exceeded 10%, making them competitive candidates toward large-area and low-cost light-emitting applications allowing printing techniques. Similar to other LED categories, light out-coupling efficiency is an important parameter determining the EQE of perovskite LEDs (PeLEDs), which, however, is scarcely studied, limiting further efficiency improvement and understanding of PeLEDs. In this work, for the first time, optical energy losses in PeLEDs are investigated through systematic optical simulations, which reveal that the 3D and quasi-2D PeLEDs can achieve theoretically maximum EQEs of $\approx 25\%$ and $\approx 20\%$, respectively, in spite of their high refractive indices. These results are consistent with the reported experimental data. This work presents primary understanding of the optical energy losses in PeLEDs and will spur new developments in the aspects of device engineering and light extraction techniques to boost the EQEs of PeLEDs.

Organic–inorganic perovskite materials have attracted extensive interest due to their fascinating semiconducting properties, such as high absorption coefficient, long carrier diffusion length, and small exciton binding energy.^[1–3] Moreover, these materials are easily processed via solution-based techniques, showing great potential applications in low-cost optoelectronic devices such as solar cells,^[1–3] light-emitting diodes (LEDs),^[4–7] lasers,^[6–8] and transistors.^[9–11] In the past few years, we have witnessed the rapid improvement of power conversion efficiency of perovskite solar cells, which reaches over 22% in 2016.^[12]

In addition to their excellent properties for light absorbers, perovskite materials are also outstanding emitters with high color purity, high photoluminescent quantum yield (PLQY), and

easily tunable band gaps.^[4–7] LEDs based on such materials have been attracting increasing attention since the first room-temperature perovskite LEDs (PeLED) developed in 2014.^[13] The development of PeLEDs has progressed very rapidly, realizing a record-high external quantum efficiency (EQE) of 11.7% in 2016.^[14]

Similar to other LED categories, e.g., organic LEDs (OLEDs)^[15] and quantum-dot^[16,17] LEDs, the EQE of PeLEDs is determined by the internal quantum efficiency (IQE) and light out-coupling efficiency (η), which is expressed as

$$\text{EQE} = \eta \cdot \text{IQE} = \eta \cdot \gamma \cdot \eta_{\text{S/T}} \cdot q_{\text{eff}} \quad (1)$$

The IQE is the product of the charge carrier balance (γ), the fraction of excitons capable of radiative decay at room temperature ($\eta_{\text{S/T}}$), and the effective radiative quantum yield (q_{eff}). For 2D perovskite materials with relatively large binding energy, the $\eta_{\text{S/T}}$ could be unity when heavy atoms are involved in the organometallic complex (similar to that in phosphorescent materials^[18]). For 3D perovskite materials with relatively small binding energy, the emission should be band-to-band transitions of free carriers in direct gap semiconductors, and the $\eta_{\text{S/T}}$ term should not be involved in the equation in this case. All in all, this equation shows that the EQE is largely influenced by the η besides the IQE. Thus, studying the limits of η is of great importance for achieving high-EQE PeLEDs.

As the fast development of PeLEDs, a conflict appears between the experimental and theoretical results. In general, the η can be estimated according to the ray-optics theory,^[19]

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which has been well acknowledged in OLEDs as a first approximation, that is

$$\eta = \frac{1}{2n^2} \quad (2)$$

where n is the refractive index of the emitting material. As for PeLEDs based on methylammonium lead iodide (MAPbI₃) and NFPI₇ (the mixture of 1-naphthylmethylamine iodide, formamminium iodide, and PbI₂ with the molar ratio of 2:1:2), it would be 7.4% and 12.5% respectively because of their large refractive index of ≈ 2.6 and ≈ 2.0 , respectively, at the emissive wavelength. The η can be estimated using Equation (1) from experimental data based on the EQE and the PLQY with the assumption of good charge carrier balance ($\gamma = 1$). By using this method, a η of $\approx 19.5\%$ can be obtained for the NFPI₇-based PeLEDs.^[14] Such a conflict implies that the optical processes in PeLEDs are quite different from those in the OLEDs, and more complicated than what the ray-optics theory can predict. Therefore, more precise optical simulations are highly desirable to understand the optical energy losses in PeLEDs and predict their theoretical EQE limits.

Herein, we systematically study the optical energy losses in PeLEDs through advanced optical simulations. Our results show that the maximum η ($\eta_{\max}$) of MAPbI₃-based 3D and NFPI₇-based quasi-2D PeLEDs are $\approx 25\%$ and $\approx 20\%$, respectively. However, the 3D PeLEDs based on MAPbI₃ with relatively larger refractive index show η values that are more sensitive to the thickness of the emitting layer (EML), whereas the quasi-2D PeLEDs based on NFPI₇ with relatively smaller refractive index show less thickness dependence. In addition, there is strong coupling between the waveguide mode and evanescent/surface plasmon polariton (SPP) mode in PeLEDs with large refractive indices, resulting in new hybrid optical modes and different spatial power distribution inside the PeLEDs. These results suggest that further development of high-efficiency PeLEDs can be realized through device engineering or reducing the substrate and the hybrid optical modes using various light extraction technologies,^[20,21] e.g., attaching an index-matched

hemisphere at the air/glass interface and forming microstructures at the ZnO/EML interface.

We adopt the classical electromagnetic model to describe the electroluminescence in PeLEDs (details are described in the Experimental Section). In the classical electromagnetic model, the radiative decay of excitonic species are treated as an ensemble of classical electrical dipole antennas radiating electromagnetic power.^[22–28] The device structure used for simulations is shown in **Figure 1a**, consisting of indium tin oxide (ITO), zinc oxide (ZnO), perovskite EML, poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,4-(N-(4-sec-butylphenyl)) diphenylamine)] (TFB), molybdenum oxide (MoO₃), and gold (Au). Two widely reported green and near-infrared 3D perovskite materials,^[13,29–31] methylammonium lead bromide (MAPbBr₃) and MAPbI₃, and the highest-efficiency quasi-2D perovskite NFPI₇^[14] are chosen as the EMLs in this study. The refractive indices of MAPbI₃ and NFPI₇ are measured to be ≈ 2.6 and ≈ 2.0 at their emissive wavelengths (700–850 nm), respectively, and that is ≈ 2.2 for MAPbBr₃ at 450–550 nm (Figure 1b). The refractive indices of other materials used in this study are shown in Figure S1 in the Supporting Information. In consideration of the wide distribution of refractive indices of perovskite materials, various refractive index values (1.75–3.00) are studied.

The simulated dispersion diagrams of transverse magnetic (TM) polarized light are shown in **Figure 2**, which indicate how the radiated power changes with the energy and the in-plane wavevector. Each graph in Figure 2 is divided into three regions corresponding to the direct emission mode (Region 1, light into the air from the forward hemisphere direction), the substrate mode (Region 2, light trapped inside the glass substrate), and the waveguide/SPP mode (Region 3, light trapped inside the functional layers), respectively. The details of how to define the three regions are presented in the Experimental Section. As for the direct emission mode (Region 1) and the substrate mode (Region 2), their dispersion relation rarely changes with the refractive index of the EML. However, in Region 3, three continuous bright curves (namely TM₀, TM₁, and TM₂ in the order from the right-bottom corner to the left-top corner) appear in Figure 2a, indicating different TM modes. As the refractive

Figure 1. a) Schematic illustration of the device structure of the PeLEDs used for optical simulations. b) The refractive indices and photoluminescence spectra of the selected perovskite materials.

Figure 2. Dispersion diagrams of transverse magnetic (TM) polarized light as the function of the energy (or wavelength) and the in-plane wavevector of various refractive indices of the EML. The refractive indices of EML are a) 1.75, b) 2, c) 2.25, d) 2.50, e) 2.75, and f) 3.00.

index of the EML increases, the waveguide modes (TM_1 and TM_2) gradually shift toward the large in-plane wavevector. When the refractive index of the EML is larger than 2.5, the TM_1 curve is significantly twisted at the energy between 2.1 and 2.5 eV (corresponding to emissive wavelength between 500 and 600 nm), and tends to overlap with the TM_0 curve. This result suggests that the SPP mode and the waveguide mode begin to couple with each other to generate new hybrid TM modes. As the change of TM_0 and TM_1 modes around 760 nm (the central emissive wavelength of MAPbI₃) are not as obvious as those between 500 and 600 nm, the new hybrid TM modes do not definitely exist in PeLEDs based on perovskites such as MAPbI₃. Further investigation on the full power dissipation spectra of these devices at 760 nm (Figure S2, Supporting Information) shows the same result. When the refractive index of the EML increases, the resonant peaks of TM_0 and TM_1 modes only shift slightly toward larger in-plane wavevectors.

To further study the coupling between the SPP mode and the waveguide mode in the PeLEDs with the infrared emission, we investigate the spatial distribution of power density of TM_0 and TM_1 modes (at 760 nm) inside the devices where the EMLs have different refractive indices. As shown in Figure 3a, where the refractive index of the EML is 1.75, the power intensity of TM_0 decreases exponentially at the MoO₃/Au interface. This is the critical feature of the SPP mode.^[32] The profile of TM_1 is a typical sinusoid shape of the waveguide mode,^[33] which goes across the glass/ITO/ZnO layers in Figure 3a. Figure 3a,b shows bare coupling between the TM_0 and TM_1 modes. However, when the refractive index increases to 2.25 and above (Figure 3c–f), one sinusoid wave across the EML appears in the TM_0 curves. This observation indicates that the SPP mode couples with the waveguide mode in the PeLEDs with refractive index of >2.25. Besides, the TM_1 curves in Figure 3c–f shows the feature of SPP mode at the MoO₃/Au interface. Therefore,

strong coupling between the waveguide mode and the SPP mode does exist in infrared PeLEDs (i.e., MAPbI₃-based devices), resulting in two new hybrid TM modes and distinctive spatial power distribution. From the perspective of the light extraction, one nanostructure at the ZnO/EML interface could be very efficient to extract light trapped by both the waveguide and the SPP modes at the same time for the PeLEDs with large- n EML because both modes have high power density near this interface.

To quantitatively analyze how the refractive index of the EML affects the η , the power distribution into different optical modes in the PeLEDs are calculated through the transfer matrix approach of a classical electroluminescent model. Figure 4 depicts the simulated power distribution in different optical modes of PeLEDs with various EMLs whose refractive indices change from 1.75 to 3.00. As demonstrated above, the waveguide mode could couple with the SPP mode when the EML has high refractive index. Therefore, the power fraction of the waveguide and SPP modes are added together. In Figure 4a, where the refractive index of the EML is 1.75, the η hardly changes with the thickness of the EML, and shows its maximum at $\approx 16.7\%$. This value is consistent with the theoretical calculation according to the ray-optics theory in LEDs based on organic emitters whose refractive index is ≈ 1.7 .^[20,34,35] Interestingly, the $\eta_{\max}$, which is supposed to be smaller according to the ray-optics theory, does not reduce as the refractive index of the EML increases (Figure 4b–f). In contrast, it fluctuates around 17%, and the $\eta_{\max}$ even increases to 25.1% when the refractive index reaches 2.50 or above (Figure 4d–f). It is also interesting to note that larger refractive index makes the η values more sensitive to the thickness of the EML, ranging between 11.5% and 25.1% when the refractive index is 2.50 (Figure 4d). Therefore, the large refractive index of perovskite materials will strengthen the interference of light inside the thin films.

Figure 3. Spatial distribution in every functional layer of the power density of TM_0 and TM_1 modes at 760 nm. The refractive indices are a) 1.75, b) 2.00, c) 2.25, d) 2.50, e) 2.75, and f) 3.00. The thicknesses of the EMLs used in the simulations are those resulting in the maximum light out-coupling efficiency, which are a) 230, b) 210, c) 190, d) 170, e) 150, and f) 130 nm, respectively.

It will directly affect the power fraction of waveguide and SPP modes. However, through careful design of the device structure (optimization of the thickness of each layer), a high η of $\approx 25\%$ can still be achieved.

In Figure 4 and related discussion, the refractive index of the EML is assumed constant over the emissive region. The power distribution into different optical modes in PeLEDs based on the real refractive indices of $MAPbI_3$ and $NFPI_7$ are presented in Figure 5. The $\eta_{\max}$ values of $\approx 25.7\%$ and $\approx 19.6\%$ can be achieved in devices based on $MAPbI_3$ and $NFPI_7$, respectively. The latter is very close to the estimation value (19.5%) from the experimental results.^[14] This simulating result indicates that PeLEDs based on 3D perovskites (i.e., $MAPbI_3$) show relatively higher $\eta_{\max}$ values than those based on quasi-2D ones (i.e., $NFPI_7$). However, the η values of the 3D PeLEDs are more sensitive to the thickness of the EML (Figure 5a), suggesting that optimization of the thickness of the EML will not only influence the charge transport balance in the device but also change the η values. Therefore, a compromise between the charge transport balance and the η value should be taken into consideration

to achieve high EQEs in 3D PeLEDs. In contrast, for quasi-2D PeLEDs, the η value is hardly sensitive to the thickness of the EML (Figure 5b), indicating that the charge transport balance should be the primary concern when doing device engineering. From the observations of Figures 4 and 5, we can conclude that PeLEDs based on 3D perovskites with relatively larger refractive indices will have higher $\eta_{\max}$ values than those based on quasi-2D perovskites with smaller refractive indices. However, the η values of 3D PeLEDs are highly dependent on the thickness of the EML, whereas they are almost invariant in quasi-2D PeLEDs.

The recombination zone of a LED could be located at either side of the EML. In addition to the above-mentioned case of TFB/EML interface, we also study the optical energy losses in PeLEDs when the recombination zone is located at the ZnO/EML interface. As shown in Figures S3 and S4 in the Supporting Information, the EQE variation with the thickness of the EML becomes larger for the cases based on smaller refractive index material and $NFPI_7$. This is because the distance between the recombination zone and the metal electrode changes with the thickness of EML as well, which would arise serious optical destructive interference.

In conclusion, we have systematically studied the optical energy losses in PeLEDs by optical simulations and demonstrated how the large refractive index of perovskite materials influences the optical losses. The simulation results show that the refractive index of the EML does affect the light out-coupling efficiency η . However, high $\eta_{\max}$ values of $\approx 25\%$ can be achieved in $MAPbI_3$ -based 3D PeLEDs through careful device engineering. For $NFPI_7$ -based quasi-2D PeLEDs, relatively lower $\eta_{\max}$ values of $\approx 20\%$ can be achieved. Our results also suggest that a compromise between the charge transport balance and the η value should be taken into consideration to achieve high-EQE 3D PeLEDs, because the larger refractive index will make the η more sensitive to the thickness of the EML. However, for quasi-2D PeLEDs with relatively smaller refractive indices, the charge transport balance should be the primary concern when doing device engineering. In addition, we find that new hybrid TM modes appear and the different spatial power distribution could make it easier to enhance the η through light extraction techniques. For example, it may be helpful to make the perovskite layer into nanostructures during its crystallization in the perspective of light extraction. Our results present primary understanding of the optical energy losses in PeLEDs and will spur new developments in the aspects of device engineering and light extraction techniques to achieve high-efficiency PeLEDs.

Experimental Section

Simulation Model: A classical electromagnetic model was adopted, treating the radiative decay of excitonic species as an ensemble of classical electrical dipole antennas radiating electromagnetic power. This was widely used in the simulation of the spontaneous emission inside a structure composed of stacked thin films.^[22–27,35–38]

In the modeling, the following assumptions were made:

- 1) The dipole orientation of emitters is isotropic. Isotropic transition dipole moment is a common case in the discussion. The total radiated

Figure 4. Power distribution in different optical modes of PeLEDs with various refractive indices of the EML. The refractive indices are a) 1.75, b) 2.00, c) 2.25, d) 2.50, e) 2.75, and f) 3.00. The recombination zone is located at the EML/TFB interface. The emissive spectrum is the same as that of MAPbI₃.

power has a linear relationship with the fraction of the horizontal/vertical dipoles as indicating by the following equation

$$P_{\text{total}} = \alpha P_{\text{horizontal}} + (1 - \alpha) P_{\text{vertical}} \quad (3)$$

where α is the fraction of horizontal dipoles, which equals to 2/3 in the isotropic case. In recent studies,^[39–42] the characterization of the angular dependent *p*-polarized PL emission spectra of the single emitting layer on a quartz substrate has been adopted to determine the orientation of the transition dipole moments. Higher out-coupling efficiency can be expected, if one perovskite material has more horizontal emitters.

- 2) The properties of all materials are isotropic. This is an intrinsic limitation of the electromagnetic model we use in this work. To deal with anisotropic materials, an effective refractive index is commonly used and it can be expressed as follows for uniaxial cases^[43]

$$\frac{1}{n_{\text{eff}}^2} = \frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2} \quad (4)$$

where θ is the angle between the propagation direction and the normal of the surface.

- 3) The emitting medium is nonabsorbing at the emissive region. This is also an intrinsic limitation of the electromagnetic model. Complex-valued refractive indices could result in numerical divergences in the calculation. For perovskite materials which have large Stocks-shift (i.e., weak overlap between the absorption spectrum and the luminescent spectrum), for example NFPI₇, the assumption is valid^[14]; For other perovskite materials, the description of the reabsorption effect can be included in the photoluminescent quantum efficiency. Because excitons from reabsorption will eventually decay through either radiative or nonradiative process. Therefore, it would not affect the calculation results of out-coupling efficiency seriously and can be treated

Figure 5. Power distribution in different optical modes of PeLEDs based on optical parameters of a) MAPbI₃ and b) NFPI₇. The recombination zone is located at the EML/TFB interface.

independently. A similar case is using the electromagnetic model to simulate the quantum-dot LEDs,^[38] while quantum dots usually have small Stokes-shift.

- 4) A discrete emitter position is used, i.e., an infinitely narrow distribution at the interface between the emitting layer and the charge-transporting layer. This is a common case due to the present lack of specific information for the distribution profile of emitters.
- 5) The devices are operated at low excitation levels or at low exciton concentrations in the emitting layer. This means exciton annihilation processes, e.g., Auger recombination, are not included in the discussions. Because relative physical models are unavailable or still under development. And this assumption can be satisfied in all real devices.

To obtain solutions of electromagnetic fields or power densities, two numerical calculation methods were adopted considering the time and data amount of the calculation tasks. Both of methods were well reported and widely used. First, developed for modelling of multilayer organic light-emitting diodes, a transfer matrix approach to describe radiating dipoles located in the emission layer was used, which was well described by Furno et al.^[28] It was very convincing in the quantitative analysis of the radiated power from the dipoles.^[28,36–38,44] Another numerical calculation based on the finite-difference time-domain (FDTD) method^[45–47] was implemented. It was used in the analysis of the dispersion relation and spatial power distribution of various optical modes.^[48–52] In the FDTD model, the boundary conditions perpendicular and parallel to the interfaces were Bloch and perfect matching layers. The simulation time step was 0.00233507 fs and the simulation time was 1000 fs. The mesh step was 0.5 nm over the device region and auto nonuniform mesh was used over other regions. An electric dipole was used as the light source.

Optical Modes in the Dispersion Diagrams: The in-plane wavevector in the medium is

$$k_{||} = k_0 \cdot n \cdot \sin\theta \quad (5)$$

where $k_0 = 2\pi/\lambda$ is the wavevector in the vacuum. θ is the angle between the wavevector and the normal of the interface. As $0 \leq \theta \leq \frac{\pi}{2}$, each diagram in Figure 2 can be divided into three regions according to different refractive indices by the color lines. The red dot and green dash lines in Figure 2 are respectively corresponding to the maximum in-plane wavevector in the air ($k_{||} = k_0 \cdot n_{\text{air}}$) and the glass substrate ($k_{||} = k_0 \cdot n_{\text{glass}}$). Thus, Region 1 is the direct emission, which is the area between the vertical axis and the red dot line ($0 \leq k_{||} \leq k_0 \cdot n_{\text{air}}$). This region represents the fraction of photons that can escape into the air from the forward hemisphere direction. Region 2 is the substrate mode, which is between the red dot line and the green dash line ($k_0 \cdot n_{\text{air}} \leq k_{||} \leq k_0 \cdot n_{\text{glass}}$). It shows the fraction of photons trapped inside the device due to the total internal reflection at the air/glass interface. The left ($k_0 \cdot n_{\text{glass}} \leq k_{||} \leq k_0 \cdot n_{\text{eff}}$) is Region 3 representing the waveguide mode and the evanescent/SPP mode. This region demonstrates photons trapped inside the thin films. The bright and dark regions denote the high and low power density, respectively.

Preparation of Perovskite Thin Films: NFPI₇ precursor solutions were prepared by dissolving 1-naphthylmethylamine iodide (NMAI), formamidinium iodide (FAI), and PbI₂ with a molar ratio of 2:1:2 in dimethylformamide (DMF) (10 wt%) and stirred at 60 °C for 2 h in a nitrogen-filled glovebox. The NFPI₇ film was prepared by spin-coating the precursor solution onto a Si substrate at 4000 rpm for 30 s, followed by annealing on a hot plate at 100 °C for 10 min in the nitrogen-filled glovebox. The MAPbI₃ precursor solution was synthesized by mixing MAI and PbI₂ in a 3:1 molar stoichiometric ratio in DMF to obtain a 0.5 M solution. The solution was spin-coated on a clean Si substrate at 4000 rpm for 30 s, to which 0.4 mL toluene was dropped after 10 s of spinning, followed by annealing on a hot plate at 100 °C for 10 min. The MAPbBr₃ film was prepared in the same way as MAPbI₃.

Characteristics: The refractive indices and thicknesses used in the simulations were measured by a variable angle spectroscopic ellipsometer (J. A. Woollam Co., Inc.). The spectra were collected at 35°, 45°, 55°, and 65°. And all samples were spin-coated on the Si wafer and all spectra were fitted using the model of “Si with absorbing film.”

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.

Keywords

external quantum efficiency, light out-coupling efficiency, light-emitting diodes, optical energy loss, perovskites

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