TOPICAL REVIEW

Light extraction from organic light emitting diodes (OLEDs)

Ruth Shinar$^1$ and Joseph Shinar$^2$

$^1$Microelectronics Research Center and Electrical & Computer Engineering Department, Iowa State University, Ames, IA 50011, United States of America
$^2$Ames Laboratory - US Department of Energy and Physics & Astronomy Department, Iowa State University, Ames, IA, 50011, United States of America

E-mail: rshinar@iastate.edu and jshinar@iastate.edu

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Abstract

Organic light emitting diode (OLED) technology continues to make strides, particularly in display technology, with costs decreasing and consumer demand growing. Advances are also seen in OLED solid state lighting (SSL) though broad utilization of this technology is lagging. This situation has prompted extensive R&D to achieve high-efficiency SSL devices at cost-effective fabrication. Here we review the advances and challenges in enhancing forward light outcoupling from OLEDs. Light outcoupling from conventional bottom-emitting OLEDs (through a transparent anode) is typically ~20%, largely due to external losses, i.e., substrate waveguide modes, internal waveguide modes between the metal cathode and the anode/substrate interface, and surface plasmon-polariton modes at the metal cathode/organic interface. We address these major photon loss paths, presenting various extraction approaches. Some approaches are devoid of light extraction structures; they include replacing the commonly used ITO anode, manipulating the refractive index of the substrate and/or organic layers, and evaluating emitters with preferential horizontal transition dipoles. Other approaches include the use of enhancing structures such as microlens arrays, scattering layers and patterned substrates, as well as substrates with various buried structures that are planarized by high index layers. A maximal external quantum efficiency as high as 78% was reported for white planarized OLEDs with a hemispherical lens to extract the substrate mode. Light outcoupling from OLEDs on flexible substrates is also addressed, as the latter become of increasing interest in foldable displays and decorative lighting, with plastic substrates also being evaluated for biomedical, wearable, and automotive applications.
## Contents

1. Introduction .......................................................... 3
2. Substrate mode extraction ........................................... 5
   2.1. Microlens arrays (MLAs) and macro (hemispherical) lens ............ 5
   2.2. Air-side scattering structures ...................................... 7
   2.3. Index-matching fluids (IMFs) ..................................... 8
3. Minimizing internally waveguided light and SPPs .................... 8
   3.1. Enhanced light extraction with no extracting structures ............... 9
      3.1.1. ITO-free OLEDs ............................................. 9
      3.1.2. Manipulating the refractive index of the organic layers ........... 9
      3.1.3. OLEDs with preferential horizontal transition dipole moments 10
   3.2. OLEDs with light outcoupling enhancing structures .................. 11
      3.2.1. OLEDs on high index substrates ................................ 11
      3.2.2. OLEDs on photonic crystal (PC) structures ........................ 11
      3.2.3. Scattering layers ............................................ 12
      3.2.4. Patterned and planarized OLEDs .............................. 13
4. OLEDs on flexible substrates ....................................... 19
5. Summary and concluding remarks ................................... 24
Data availability statement ........................................... 24
Acknowledgments ....................................................... 24
References .............................................................. 24
1. Introduction

This review highlights various approaches and advances, as well as ongoing challenges, in enhancing light outcoupling from organic light emitting diodes (OLEDs). It is not intended to be a comprehensive survey of the wide-ranging reports in this broad area and will hence provide only representative examples.

OLEDs continue to make impressive strides in applications such as displays and solid state lighting (SSL). While their use in TVs and other displays is expanding, with prices steadily decreasing, wide OLED usage in SSL is lagging, with devices being expensive and often attractive mostly for niche applications, such as flexible and decorative light fixture designs. In addition to cost, an ongoing related impediment leading to this situation is the issue of OLED efficiencies and the light outcoupling or extraction factor $\eta_{\text{out}}$, which is the fraction of photons emitted into the forward (viewing) hemisphere. Earlier reviews have addressed different aspects of this issue [1–7]. The external quantum efficiency $\eta_{\text{EQE}}$, i.e. the ratio of such forward extracted photons to injected electrons, is given by

$$\eta_{\text{EQE}} = \eta_{\text{IQE}}\eta_{\text{out}}$$  \hspace{1cm} (1)

where $\eta_{\text{IQE}}$ is the internal quantum efficiency, i.e. the ratio of photons generated in the device to electrons injected into it. $\eta_{\text{IQE}}$, in turn, is given by

$$\eta_{\text{IQE}} = \gamma r_{\text{em}}\eta_{\text{PL}}$$  \hspace{1cm} (2)

where $\gamma (\leq 1)$ is the charge balance factor, $r_{\text{em}} (\leq 1)$ is the fraction of excited states, generated by electron–hole recombination, which are emitting states (typically 0.25 for promptly fluorescent singlet and 1.00 for phosphorescent triplet materials), and $\eta_{\text{PL}} (\leq 1)$ is the photoluminescence (PL) quantum efficiency.

In general, for standard through-the-transparent-bottom-emitting OLEDs, devoid of any light extraction structures, $\eta_{\text{out}}$ is approximated by [8, 9]

$$\eta_{\text{out}} = 1 - \sqrt{1 - n_{\text{org}}^{-2}} \approx 0.50/n_{\text{org}}^2 \sim 0.2$$  \hspace{1cm} (3)

where $n_{\text{org}} \sim 1.70–1.75$ is the refractive index (RI) of the emitting layer (EML). However, equation (3) is valid for randomly oriented emitting dipoles (anisotropy factor $\alpha_{\text{an}} = 0.33$) not subjected to optical interference (i.e., cavity effects) with the reflecting cathode [10, 11]. If such optical interference effects are taken into account, then [9]

$$\eta_{\text{out}} \approx 0.75/n_{\text{org}}^2 \sim 0.25.$$  \hspace{1cm} (4)

If the dipoles are all in the plane of the OLED ($\alpha_{\text{an}} = 0$) and optical interference with the cathode is taken into account, then [9–11]

$$\eta_{\text{out}} \approx 1.2/n_{\text{org}}^2 \sim 0.40.$$  \hspace{1cm} (5)

As with randomly oriented emitting dipoles $\eta_{\text{out}} \sim 0.2–0.25$; ~75%–80% of the photons are lost to various processes, mostly the following:

(a) Light externally waveguided in the substrate (substrate mode) due to total internal reflection (TIR) at the substrate/air interface. Technically, they are waveguided between the reflecting Al cathode and the glass/air interface. However, as the organic layers + ITO are only ~200 nm thick, whereas the glass substrate is ~1 mm thick, effectively they are waveguided in the glass.

(b) Light internally waveguided in the high index organic layers ($n_{\text{org}} \sim 1.75$) and anode, typically ITO [$n_{\text{ITO}}(700 \text{ nm}) = 1.82, n_{\text{ITO}}(410 \text{ nm}) = 2.10$].

(c) Formation of surface plasmon polaritons (SPPs) at the organic/metal cathode interface in single stack devices. This issue diminishes in multi-stacked, tandem devices, as this mechanism weakens rapidly with increasing distance between the EML and the cathode.

Notwithstanding the foregoing loss mechanisms, there are reports of $\eta_{\text{EQE}}$ exceeding 25% with no added external extracting structures. For example, efficient green phosphorescent OLEDs with a maximal $\eta_{\text{EQE}} = 29.1\%$ were demonstrated using an EML with an exciplex forming co-host [12]. In other examples, enhanced efficiencies were achieved in green and blue OLEDs with a poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) anode [13]. Similarly, enhanced outcoupling was observed in green ITO-free fluorescent [14] and phosphorescent OLEDs with multilayered PEDOT:PSS anodes [15]. These performance enhancements were attributed to the advantageous RI of anodes, such as PEDOT:PSS.
(\(n_{\text{PEDOT}} \sim 1.5\)), which is closely index matched to glass (\(n \sim 1.5\)), unlike ITO (\(n \sim 1.8–2.1\)), which results in significant photon loss due to TIR at the ITO/glass interface.

Figure 1 presents the main loss processes listed above in standard bottom emitting OLEDs [16] and figure 2 shows these and other loss processes in a red-emitting device [17]. The photon loss channels, as predicted by optical modeling, are depicted in the figure as a function of the electron transport layer (ETL) thickness; experimental EQE values (circles) are also shown. As expected, \(\eta_{\text{out}}\) is only \(\sim 20\%\) due mostly to the loss mechanisms mentioned above. As seen in figure 2, the SPP loss decreases significantly as the ETL thickness increases but the internally waveguided loss increases; generally, the latter two losses amount to \(\sim 50\%\) of the photons. As the thickness of the ETL, and in particular, as the number of units increases, the SPP-related loss becomes less significant, and hence current industrial efforts to enhance OLED SSL are focused on optimally reducing the internally waveguided loss channel, and extracting the externally waveguided light, trapped in the substrate.

The dependence of the above-mentioned loss processes on the ETL thickness is further shown in figure 3 for phosphorescent red and green, as well as for fluorescent blue OLEDs [18]. An optical microcavity effect between the Al and ITO electrodes results in a standing wave in which there are antinodes at the EML and at

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**Figure 1.** Photon loss paths in standard bottom emitting OLEDs. Reproduced from [16] with permission from the Royal Society of Chemistry.

**Figure 2.** Optical modeling of the EQE vs ETL thickness for a red emitting OLED. The dark circles show experimental data. Reproduced from Meerheim et al [17], with the permission of AIP Publishing.
the ITO/glass interface and a node at the Al electrode. Hence, the minimal distance between the EML and at the ITO/glass interface is \( \lambda/(2n_{\text{org}}) \) nm, and the Al is separated by \( \lambda/(4n_{\text{org}}) \) nm from the EML, which is \( \sim 75 \) nm for a green OLED (assuming \( n_{\text{org}} \sim 1.75 \)).

Various approaches have been developed in an attempt to achieve the goal of increasing \( \eta_{\text{out}} \) from \( \sim 20\% \) in standard bottom emitting OLEDs to >70%. As presented below, the approaches include extracting the substrate mode and the internally waveguided mode, as well as minimizing the SPP loss, though some of the approaches address simultaneously more than one photon loss path. As shown, the reduction of the substrate and internally waveguided modes requires reducing TIR via e.g., control of the RI of the various layers in the OLEDs, as well as scattering and diffracting designs. As SPP formation is significant at the smooth organic/metal cathode interface, patterning that interface or increasing the distance of the EML from the cathode, as in tandem structures, minimizes that loss. Design principles of the various approaches that decrease TIR-related losses and minimize SPP formation are also provided in the different sections to elucidate the role in light extraction enhancement of the approach.

The following section covers substrate mode light extraction. This is followed by sections on extracting internally waveguided light and mitigating the SPP loss. Included are flat, patterned, and planarized devices on glass and plastic. We note that division into different sections cannot be strict, as many studies employed various enhancing approaches simultaneously. The significant examples highlighted within each section typically start with earlier reports and advance toward recent ones. In addition to presenting various approaches, including state of the art, ongoing challenges are also addressed.

2. Substrate mode extraction

Approaches to extract the substrate mode vary, with microlens arrays (MLAs) possibly the most widely used [19]. Other approaches include the use of a hemispherical lens, surface texturing and high RI nanoparticle-and/or scattering micro-particle doped layers, either coated on the substrate or embedded in it [1–5, 20–28]. These approaches, like others, are often combined with additional extraction means for minimizing the other loss processes. Scattering layers currently remain among the most promising structures for enhancing outcoupling in e.g., commercial SSL. The addition of substrate mode extraction structures often eliminates the dependence of the EL on the wavelength and viewing angle via scattering, as can be the case with some outcoupling enhancing diffractive structures, for example.

2.1. Microlens arrays (MLAs) and macro (hemispherical) lens

MLAs fabricated by various techniques using different materials are commonly and successfully used at the glass/air interface of bottom-emitting devices [2]. They have been utilized in different applications, including for enhancing solar cells and OLEDs, as well as in optical sensors [29]. Application of MLAs to top emitting devices requires modified approaches to avoid damage to the underlying organic layers, though theory predicts a significant enhancement [30].
Via multiple reflections, MLAs reduce the incident angle below the critical value at the substrate/air interface, decreasing the TIR loss and increasing the EQE. On average, light outcoupling enhancements of $\sim 1.5 \times$ to $\sim 1.7 \times$ with different MLA or prism designs, some complex, were achieved. A MLA in conjunction with standard bottom-emitting devices may be more suitable for some industrial applications than a hemispherical lens, where an outcoupling enhancement $> 2 \times$ was demonstrated [2, 31].

In an early report, Sun and Forrest [32] studied the dependence of the MLA geometry, specifically the microlens diameter, the height/pitch ratio, and the microlens-to-OLED diameter ratio, on $\eta_{\text{out}}$ in Al/organic/ITO/glass OLEDs. Their simulations showed that for a 1 mm OLED and a hexagonal array of microlens touching each other and making a contact angle of $70^\circ$ with the substrate, $\eta_{\text{out}} \approx 31\%$, independent of the microlens-to-OLED diameter ratio. Their simulations also show that the outcoupling enhancement increases with increasing microlens diameter up to $\sim 2 \mu m$, and remains constant beyond that value. In these simulations, the ratio of the microlens height to the pitch was close to 0.5.

In one example, an enhancement of $\sim 2 \times$ in the electroluminescence (EL) intensity was obtained with a MLA area that is significantly larger than that of the OLED pixel [33]. Figure 4 shows this situation for green and blue fluorescent OLEDs with pixel areas of $3 \times 3 \text{ mm}^2$ and MLA area of $16 \times 16 \text{ mm}^2$. The MLA structure had a $2 \mu m$ pitch, $1.6 \mu m$ diameter, and $1.2 \mu m$ height; it was fabricated directly on the air-side of the OLED's glass substrate by imprinting a polydimethyl siloxane (PDMS) mold on a polyurethane (PU) film prior to OLED fabrication. The master mold was fabricated by laser interference lithography. The figure also shows the energized OLED pixels with and without the MLA. The OLEDs were simply encapsulated by the bottom glass substrate, a top glass slide, and a vacuum epoxy seal around the edges. As seen, in the absence of a MLA, light that is waveguided to the edges of the substrate is seen through its scattering by the epoxy; this light is absent in the presence of the MLA.

Interestingly, Peer et al [34] showed that the foregoing MLAs [33] also enhanced the absorption and photocurrent (by 6.3%) and hence the EQE of methyl ammonium Pb halide organic/inorganic hybrid perovskite solar cells.

Recently, MLAs that were shown to enhance blue, green, and red OLEDs were formed by nano-imprinting PETs using a stainless steel stamp directly structured by laser interference patterning [35].
In other recent developments, Han et al [36] addressed the effect of the MLA outcoupling enhancement on large area emitters (up to 6084 mm²). In addition to the area, they also evaluated the viewing angle effects. They concluded that the luminance enhancement using the MLA was dependent on the viewing angle and emitting area, with smaller panels showing a relatively lower enhancement. That is, in their studies, in the normal direction the luminance enhancement exceeded 60% for 2500 and 6084 mm² panels, while it was 48% and 58% for 2.25 and 70 mm² panels, respectively.

With the expanding development of flexible and foldable optoelectronic devices, including wearable devices, the interest in flexible MLAs is growing. To that effect, unconventional flexible dual MLAs in PDMS were shown to be effective in enhancing OLED light intensity in comparison to more conventional structures [37].

Recently, Ding et al reported the fabrication of flexible PDMS touch-screen protector films with MLAs of different parameters that enhance the brightness of an OLED by over 14% [38]. These flexible screen protectors with MLAs are promising for flexible and foldable optoelectronic devices.

In addition to MLAs, hemispherical lenses have also been employed to extract the substrate mode. If the hemispherical lens, with a RI approximately equal to that of the substrate, is much larger than the OLED pixel, virtually no photons will be trapped in the substrate because all of them will be incident on the hemispherical lens/air interface at nearly normal incidence. In an early demonstration of its effect, EQE_max of green 4,4′-N,N′-dicarbazole-biphenyl:bis(2-phenylpyridine)(acetylacetone)iridium(III) (CBP:Ir(ppy)_2(acac))-based OLEDs [39] increased from 29.2% without the lens to 54% with the lens, an addition of 24.8% and an enhancement factor of 1.85. However, it should be emphasized that while the large hemispherical lens does indeed extract nearly all of the light trapped in the substrate mode, for obvious reasons its commercial value is problematic.

2.2. Air-side scattering structures
Air-side scattering structures, such as MLAs and half spheres also successfully extract the substrate mode [2–5, 20, 24–28] via scattering and TIR reduction. Peng et al [20] used anodized alumina as the substrate. The rough anodized surface served the same function as a MLA, and the OLED was fabricated on the smooth backside of the film. And although the efficiencies of the copper phthalocyanine (CuPc)/N,N′-Bis(3-methylphenyl)-N,N′-diphenylbenzidine (TPD)/TPD:5 wt.% rubrene/tris-(8-hydroxyquinoline) aluminum (Alq3)/LiF/Al OLEDs was not measured, for viewing angles up to 60° the anodized alumina substrate increased the EL intensity >2× relative to the same OLED on standard glass/ITO.

Scattering films were also successfully utilized. In an early report [29], an ~8 µm polystyrene film embedded with 360 nm TiO2 particles (commonly used in large amounts in paints) was attached to the air-side of a glass substrate. A significant enhancement in the EL of green OLEDs was observed, though this approach was used primarily to enhance PL signals of organic electronics-based optical sensors. In a distinct and very simple approach toluene, solutions of blended polystyrene and polyethylene glycol (PS:PEG) at various weight ratios (e.g., 4:1 and 9:1) were drop cast on the air-side of a glass substrate on which an OLED was fabricated [40]. In such blended films, during the course of solvent evaporation, the PS dries faster than the PEG. The film shrinks as it dries and air voids form with the PEG decorating the PS pores [41–46], as shown in figure 5. The figure also shows a SEM image of a microporous film formed by this process and the obtained ~60% enhancement in the EL intensity [40].

The successful use of other particles was demonstrated by Song et al [26]. They fabricated orange bis(2-(3,5-dimethylphenyl)-4-phenylpyridine) Ir(III) (2,2,6,6-tetramethylheptane-3,5-diketonate) [Ir(dmpyp-ph)2tmd]-based OLEDs on ITO, i.e., with preferentially horizontal dipole emitters, and a very thick (nearly 300 µm) external scattering layer composed of SiO2 particles embedded in a Norland NOA73 host, achieving an EQE of 56%. Their simulations also confirmed that this is the maximal EQE for such OLEDs. Another relatively simple approach [28] involved a light extraction layer formed by drop casting a TCTA and 4,6-Bis(3,5-di(pyridin-4-yl)phenyl)-2-methylpyrimidine, 4,6-Bis(3,5-di-4-pyridinylphenyl)-2-methylpyrimidine (B4PyMPM) mixed solution followed by UV curing, which yields films with irregular microstructure. This scattering layer yielded blue phosphorescent OLEDs (PhOLEDs) with a maximal EQE of 44.3%.

Recently, Choi et al [47] reported enhanced light outcoupling by facile fabrication of controlled SnOx nanocones that resulted in strong scattering, reducing the TIR in the glass substrate. A maximal EQE of 44.3%, or 35% enhancement, was achieved for a blue OLED. As stated by Choi et al, this approach may be suitable for large area SSL applications on glass or flexible substrates.

Additional examples of light extraction enhancement using multi-functional scattering films, i.e., for extraction of both the external and internal waveguided photons are summarized in the next sections.
2.3. Index-matching fluids (IMFs)

A common laboratory method to extract virtually all of the photons trapped in the substrate is to couple the OLED to the photodetector, commonly a Si photodiode, with an IMF whose RI matches that of the substrate. Then almost all of the photons that penetrate the substrate continue into the IMF, and since the real part of the RI of Si is $\sim 4$ in the visible range, they are absorbed by the photodiode. Hence, comparing the EQE of an OLED with and without an IMF immediately reveals the fraction of photons lost in the substrate mode in the IMF’s absence. Since an IMF has significant diagnostic but little commercial value, it is noteworthy that to the authors’ knowledge the only methods demonstrated to date that extract essentially all of the photons in the substrate mode are the large hemispherical lens or the IMF. Hence, this indicates that extracting essentially all of the substrate mode photons remains a challenge.

3. Minimizing internally waveguided light and SPPs

Recovering the $>50\%$ internally waveguided light in the organic + anode layers + SPP losses at the metal cathode, in particular in a cost-effective, industrial approach, remains challenging. Different approaches to extract the internally waveguided light have been developed over the years; Zou et al. [48] provided a recent survey of these approaches, and hence, only some examples are provided below. It should be noted, however, that some of the approaches include complex designs that are expected to be costly, in particular for upscaling.
3.1. Enhanced light extraction with no extracting structures

There have been significant advances in developing successful extraction structures for mitigating the waveguided and SPP losses, including potentially inexpensive roll-to-roll (R2R) extracting substrates for flexible devices. These advances often rely on patterns, some planarized, resulting in preferred planar devices. The latter are further combined with MLAAs or a hemispherical lens to extract the substrate mode. Still, there are continued efforts to enhance light outcoupling via modulation of the optical properties of the devices as well [49–51], without adding light-extracting structures. These include an enhanced efficiency of top-emitting OLEDs in which SPP losses are reduced [49] and OLEDs with a low RI hole transport layer (HTL) [50] or a low RI ETL [51]. Other approaches include anode material modification and development of emitters with preferred horizontal dipole moments, which are described below.

3.1.1. ITO-free OLEDs

One approach to mitigate the problem of the high \(n_{ITO}\) anode is to replace it with the lower \(n\) e.g., PEDOT:PSS anode, to reduce TIR, as \(n_{PEDOT:PSS}\) increases from 1.49 at 700 nm to 1.54 at 400 nm. Indeed, Fehse et al [13] showed that such replacement, which involved a single ∼100 nm thick PEDOT:PSS layer, increased the power efficiency (PE) of phosphorescent red, phosphorescent green, and fluorescent blue OLEDs. The enhanced efficiency occurred in spite of the likely higher sheet resistance of PEDOT:PSS, as its conductivity was only ∼500 S cm\(^{-1}\), whereas that of ITO is ∼10\(^4\) S cm\(^{-1}\). Following that study, Cai et al [14, 15] reported a study on the effects of various PEDOT:PSS films. In particular, they described a method to deposit multilayer treated PEDOT:PSS films via spin-coating. And although the sheet resistance \(R_{sh}\) decreased with PEDOT:PSS thickness, so did the transmission at 550 nm \(T_{550}\). It was found that for phosphorescent green tris[2-phenylpyridinato-\(C_2^\text{2-}\)N]iridium(III) [Ir(ppy)_3]-based OLEDs the optimal films were 72 nm thick, 2-layer films. While the PEDOT:PSS \(R_{sh}\) (∼125 Ω/□) was much higher than that of ITO (∼12 Ω/□), and \(T_{550}\) slightly lower (88% vs 92% for ITO), the maximal EQE and PE of these OLEDs were 40% and 118 lm W\(^{-1}\), values that were higher by 63% and 44%, respectively, than those for the OLEDs on ITO. In summary, it appeared that the lower RI of PEDOT:PSS resulted in enhanced \(n_{out}\) and efficiencies by reducing the internally waveguided light.

Hecht et al [52] reviewed several transparent conducting alternatives to ITO. Of these, they noted the relatively mature and widely studied carbon nanotubes (CNTs) and CNT inks deposited by solution-processing on plastic substrates. At the same time, however, they also noted that the greatest impediment to their application as transparent electrodes was their prohibitive sheet resistance. They also reviewed films of graphene flakes and networks. While the \(R_{sh}\) of highly doped graphene is ∼62.4/\(N\) Ω/□, where \(N\) is the number of graphene layers, that of graphene networks is much higher due to their high defect density.

In a study of OLEDs on graphene, Lee et al [53] noted that while graphene-based OLEDs are very attractive for flexible displays and lighting, their efficiency is wanting. They proposed a structure in which a high-index TiO\(_2\) layer [54] and a low-index HTL sandwich the graphene anode, resulting in a strong optical cavity effect. They demonstrated EQEs up to 41% and 62% (65% and 103% with an added hemispherical lens) for single- and multi-unit (stacked tandem) OLEDs, respectively. They also demonstrated that such OLEDs on plastic were repeatedly bendable down to a 2.3 mm radius of curvature.

Considerable attention has been given to silver nanowire (AgNW)-based anodes. As an example, Spechler et al [55] described enhanced efficiency of flexible green PhOLEDs fabricated on a conductive, colorless polyimide (PI) substrate embedded with AgNWs and TiO\(_2\) particles, smoothed by a PEDOT:PSS layer. The ∼68% enhancement in the current efficiency, from ∼40 to ∼67 cd A\(^{-1}\), was attributed, among others, to the haziness of the scattering AgNW electrode, as well as to partial extraction of the waveguided light.

3.1.2. Manipulating the refractive index of the organic layers

Equation (3) shows that decreasing the RI of the organic layers increases the outcoupling factor. With this approach in mind, Salehi et al [56] demonstrated enhanced outcoupling by oblique angle deposition, which reduces the RI of the deposited layer. They showed that they could reduce the RI of the tris(8-hydroxyquinoline)aluminum (Alq\(_3\))-based ETL from 1.75 to 1.45, and consequently increase the outcoupling by 30%. Independently, Watabe et al [57] developed hole and electron injection layer (HIL and EIL, respectively) materials, based on blends of a metal oxide such as MoO\(_3\) and an organic compound, with RIs <1.6 and <1.5, respectively. As a result, their OLED EQEs increased from 30% to 41%. Clearly, the improved outcoupling is due to reduced internal waveguiding, as more photons are refracted from the organic into the ITO or other high RI anode at smaller angles, and hence more of them refract into the air. In fact, if the RI of the organic layers is reduced to that of the glass or other substrate, then almost all photons entering the anode should continue into the substrate.
3.1.3. OLEDs with preferential horizontal transition dipole moments

In a simplified picture, a dipole’s emission is strongest perpendicular to its oscillating axis. Hence, horizontal dipole orientation is favorable as the emitted light is then largely incident on the various interfaces at angles lower than the critical angle for TIR, in contrast to the case with vertically oriented dipoles that show reduced photon outcoupling and support SPP formation. However, even for horizontal dipoles, one needs to distinguish between transverse electric (TE) and magnetic (TM) polarization as the former couple weakly to SPPs but the latter couple strongly to them [58].

Many studies on guest-host OLEDs in which the transition dipole moments of the emitters are preferentially oriented have been reported [59–80]. As mentioned above, early on Kim et al [9] predicted that orienting all of the emitting dipoles in the OLED plane could increase $\eta_{out}$ from 25% to 40% as the loss to SPPs would be virtually eliminated and fewer photons would be internally waveguided. Indeed, Liehm et al [60] compared Ir(ppy)$_2$-based PhOLEDs, where the emitter dipole orientation is isotropic (anisotropy factor $a_{\alpha\alpha} = 0.33$), to Iridium(III) acetylacetonate [Ir(ppy)$_2$(acac)], where it is preferentially horizontal ($a_{\alpha\alpha} = 0.23$), and showed that the EQE increased from 18.3% to 21.7%. Similarly, Kim and coworkers [62] measured the p-polarized peak PL of Ir(ppy)$_2$(acac) in 1:1 tris(4-carbazoyl-9-ylphenyl)amine (TCTA):bis-4,6-(3,5-di-3-pyridylphenyl)-2-methylpyrimidine (B3PYMPM) at 520 nm vs emission angle. By comparing the observed behavior to simulations, they were able to show that the EQE of OLEDs that used CBP and B3PYMPM resulted in the most horizontal dipoles ($a_{\alpha\alpha} = 0.25, 0.27$), whereas UGH-2 yielded the most vertical dipoles ($a_{\alpha\alpha} = 0.40$). They attributed the horizontal preference in Ir(ppy)$_2$-tmtd to the two nearly parallel triplet transition dipole moments along the direction from the Ir core to the two nearly parallel triplet ligands with 2-fold symmetry, and the effect of the hosts on the large difference in the intermolecular binding energies. Specifically, their calculations yielded a strong ($\sim 30$ kcal/mole) binding energy of the Ir complexes with CBP and B3PYMPM, but an unstable ($\sim +6$ kcal/mole) binding energy with UGH-2. They also determined the optimized guest-host binding geometry that indeed yielded the observed preferred emitting dipole orientation.

In another follow-up development of novel heteroleptic Ir complexes, Kim et al [68] developed additional complexes where they demonstrated $a_{\alpha\alpha} = 0.135$, leading to yellow and green PhOLEDs with peak EQEs of 38% and 36%, respectively. In agreement with the foregoing discussion, they noted that this was due to the increased electrostatic interaction between the Ir complex and host molecules at the surface of the growing EML, resulting in stacking Ir complexes parallel to the film surface. Moreover, they also [67] demonstrated crystalline OLEDs based on a Pt complex emitter (peak emission at $\sim 625$ nm), with a horizontal emitting dipole ratio of 93% ($a_{\alpha\alpha} = 0.07$) and PL quantum yield (PLQY) of 96%. The OLEDs’ maximal EQE was 38.8%.

In parallel, Thompson, Forrest and coworkers [79, 80] reported extensive studies on heteroleptic Ir [79] and modifications of the molecular structure and structural templating of Pt complexes [80]. For the Ir complexes they also concluded that the presence of the acac group is responsible for the commonly measured value of $a_{\alpha\alpha} \sim 0.2$ for a large variety of (C$^\prime$N)$_2$Ir(acac) species. Specifically, they concluded that the acac ligand forms an aliphatic region on the surface of the otherwise aromatic Ir complex, and reasoned that that process on the organic surface during deposition is responsible for the observed net dipole alignment. For molecular modifications of the Pt complexes they demonstrated a $\sim 20\%$ increased fraction of horizontally aligned transition dipole moments. With the structural templating, they were able to achieve a $\sim 60\%$ increase in dipole orientation.

The effect of the transition dipole moment orientation was also studied in thermally activated delayed fluorescence (TADF) OLEDs. Lin et al [69] explored the effect of the orientation of OLEDs based on a spiroacridine-triazine hybrid, and demonstrated sky-blue OLEDs with an EQE of 37%. Ahn et al [75] demonstrated blue and deep blue TADF OLEDs, based on symmetrical and rigid oxygen-bridged boron acceptors and dipoles preferentially horizontally oriented, with maximal EQEs of 38% and 25%, respectively. Shortly thereafter, Lee et al [77] demonstrated a broad spectral range TADF blue OLEDs with EQE of 32.8%, which was a result of the high PLQY due to the choice of the host and the high horizontal transition dipole ratio of 0.76 ($a_{\alpha\alpha} = 0.24$).

In a multi-faceted approach, Huang et al [81] demonstrated an EQE of 41.5% combining a high-index Nb-doped TiO$_2$ transparent electrode of $n \sim 2.4$ together with relatively low $n$ HTL and ETL, as well as favorably horizontal dipole ($\theta_\parallel \sim 80\%$, i.e. $a_{\alpha\alpha} \sim 0.2$) emitters in blue TADF OLEDs. An EQE of 38.8% was
obtained for a PhOLED. Simulations predict further increased outcoupling efficiencies using optical modulations in optimized materials.

Finally, we note the growing commercial availability of molecules with preferentially horizontal dipole alignment.

3.2. OLEDs with light outcoupling enhancing structures
Numerous approaches, some of which are summarized next, have been suggested for enhancing light outcoupling via addition of layers/structures. These address mostly the three main loss mechanisms, i.e., the substrate mode, internal waveguiding, and SPPs, reducing TIR and SPP excitation. This section surveys representative approaches to address outstanding challenges and progress.

3.2.1. OLEDs on high index substrates
An early approach to mitigate the internal waveguide losses due to the TIR from the ITO $n_{\text{ITO}} (550 \, \text{nm}) \approx 1.9$ to the glass substrate was to use high $n$ substrates [31, 82, 83]. Indeed, using a high $n \approx 1.78$ glass substrate together with an index-matching hemisphere at the back of the substrate resulted in an EQE of 34% for a white OLED (WOLED), which was a 2.4× enhancement relative to a reference device on a low $n_{\text{glass}} \sim 1.5$ glass substrate [31]. Such substrates, however, are expensive, in particular for upscaling. Moreover, high index glass can present environmental issues (e.g., contain toxic lead).

Subsequently, Ide et al [84] at Panasonic used an outcoupling structure that was aimed at replacing expensive high- $n$ glass substrates; figure 6 shows their approach. A tandem WOLED was fabricated on the flat side of the high-index polyethylene naphthalate (PEN, $n = 1.77$) thin film, whose opposite side had an outcoupling texture; this structure was on a standard glass substrate. Together with a MLA and an anti-reflective coating, a PE of $\sim 130 \, \text{lm W}^{-1}$ and an EQE of $\sim 56\%$ at 1,000 cd m$^{-2}$ were obtained. As seen in figure 6, the high $n$ structure with the air gaps resulted in scattering that appeared to increase the outcoupling.

Notable types of sub-anode extracting structures, including scattering films, are described next.

3.2.2. OLEDs on photonic crystal (PC) structures
Other approaches include the use of substrates with optical gratings and PCs fulfilling Bragg's diffraction condition. They presented enhanced light outcoupling by mitigating the SPP loss and extracting the internally waveguided light, though they led to specific viewing directions and wavelength dependence. In studies by Do et al [85] and Lee et al [86] the OLED was fabricated on a sub-anode PC grating with SiO$_2$ ($n = 1.48$)/SiNx ($n = 1.95$); see figure 7. The figure shows that the predicted extraction enhancement is 1.5×, but the actual maximal luminous efficiency (and hence the EQE) of the fluorescent Alq$_3$-based OLEDs increased from $\sim 11.5$ to only $\sim 14.2 \, \text{cd A}^{-1}$, i.e. a 1.23× enhancement factor only.

Figure 7 also shows structured OLEDs fabricated on 2-d PCs [87]. Enhancements of 1.2× in the integrated intensity and 2.3× in the peak intensity of ITO/CuPc/NPB/Alq$_3$ devices were observed relative to devices without the PC. As mentioned above, this structure clearly mitigates the SPP loss at the organic/Al interface.

Ishihara et al [88] also fabricated enhanced OLEDs on 2-d PCs; the latter were nanoimprinted lithographically. The luminance of the OLEDs on those PCs was 1.5× that of standard devices.

Optimizing the PC grating height and period improved the outcoupling. Bocksrocker et al [89] fabricated a TiO$_2$ Bragg grating on glass/ITO by laser interference lithography and planarized it with a PEDOT:PSS layer. By adjusting the grating height and period they achieved an efficiency enhancement factor of $\sim 2$. In order to suppress the angular and spectral dependence, they added a MLA at the substrate/air interface to extract the substrate mode. This resulted in a uniform emission of the white polymer LEDs.
(PLEDs), and increased the efficiency by another factor of ∼2, so the total enhancement factor was ∼4. However, they did not provide any actual values of the efficiencies of the devices.

In a later study, also optimizing the PC grating depth and pitch, Fujimoto et al. [90] demonstrated a green OLED with an EQE of 43.4% using a 1-d PC structure directly etched into the substrate together with a hemispherical lens. SiO$_2$ was sputtered on the PC to smooth the sharp surfaces, which can lead to high leakage currents. The depth and pitch of the pattern were 125 nm and 500 nm, respectively, with the latter chosen to be close to the emission wavelength for efficient diffraction. The depth was chosen to minimize issues associated with large aspect ratios in the device layers. The authors concluded, based on simulations and experiments, that the PC reduces loss of waveguided and evanescent (i.e., surface plasmon) modes.

### 3.2.3. Scattering layers

Scattering layers for extracting the internally waveguided mode are typically fabricated by embedding scatterers, e.g., TiO$_2$ particles of suitable sizes in transparent polymer hosts. In one approach to enhance $\eta_{\text{out}}$, Chang et al. [21] deposited, via solution processing, a scattering layer between the ITO anode and the glass substrate. This layer was composed of ∼240 nm TiO$_2$ nanoparticles (NPs) [$n$(500 nm) ≈ 3.0] dispersed in EOCI130 (Everlight Chemical) polymer host matrix. The NPs were suspended and the polymer host dissolved in propylene glycol-monomethyl-ether acetate (PGMEA). The solution was spin-coated on the substrate and heat cured at 130 °C for 10 min to remove residual solvent. The solid content of the original PGMEA polymer solution was 30%, yielding 12 vol.% TiO$_2$ NPs in the final film. The authors demonstrated that the EQE of Ir(ppy)$_3$-based OLEDs increased from 20% in the reference devices (without the NP scattering layer) to 33% at 1,000 cd m$^{-2}$, i.e. a 65% enhancement with the scattering layer. Adding glass half spheres or a MLA further increased the EQE to 46%.

Pixelligent Inc. provides liquid suspensions of 5–10 nm zirconia NPs or 10–20 nm TiO$_2$ particles, both with controlled surface chemistries, to avoid particle aggregation and achieve transparent high RI ($n > 1.8$) films [91]. In addition to a high $n$ (2.2 at 500 nm), zirconia has high optical transparency from UV to infrared, high hardness, and high chemical inertness. For enhanced light extraction, it provides these suspensions with, in addition, larger scattering particles that provide a significant dielectric contrast with the high $n$-host and scatter visible light effectively. Figure 8 shows a structure, where the scattering film is between the substrate and anode. Light is reflected by the scatterers, enabling internal waveguided light and substrate mode extraction depending on the film’s $n$ relative to that of the substrate.

Using other materials, Lee et al. [92] fabricated a random light scattering layer with silver nanodots. This scattering layer was planarized with a high index ($n \sim 1.8$) layer resulting in an EQE increase of ∼×1.5 in a green PhOLED on glass. When an external light extraction layer with wrinkles was added, the EQE increased 1.67× to 36.7%.

In addition to sub-anode scattering films, enhanced scattering electrodes on plastic substrates were also reported. These will be described in some detail in section 4.
3.2.4. Patterned and planarized OLEDs

Ideally, extraction structures should be non-intrusive so that they will not adversely affect the OLEDs’ performance. Hence, they should be devoid of sharp edges, with the EL spectrum independent of viewing angle. Therefore, generally planar OLEDs with sub-anode extraction structures are preferred for achieving the above-mentioned attributes. Over the years, other types of extracting designs were reported, with some of them showing promising results.

3.2.4.1. Non-planar bottom-emitting OLEDs

Non-planar corrugated structures provide insight into the role of optics and photonics (e.g., cavity effects, TM TE, and SPP modes) in OLEDs and, as shown below, result in enhanced EQEs by minimizing the SPP loss and internally waveguided light [93–97].

On-anode structures have been fabricated and studied [98–103]. These designs included a low RI periodic structure, or high RI with scattering centers, between the anode and the organic layers. While such nanoscale structures increase the efficiency and color purity, they can adversely affect the EL leading to wavelength and angle dependence [93, 104]. Low RI microscale periodic structures, which were reported to avoid spectral distortion, showed smaller enhancement [98, 100]. For example, Sun and Forrest [99] described a square low-index SiO$_2$ grid (n$_{SiO_2}$ ~ 1.45) of 100 nm SiO$_2$ thickness and 7 $\mu$m period that eliminated grating effects. The grid was patterned on ITO, on which the high-index organic layers of a WOLED were deposited. This structure enabled extraction of the internally waveguided light into substrate and air modes, increasing the peak EQE of a standard device to ~19% from 14.7%. The addition of a PDMS MLA further increased the peak EQE to ~34% from ~25% obtained for a standard device with the MLA. This structure was found to be suitable for devices with broad spectra.

Recently, Kim et al [103] reported enhanced optical efficiency and color purity in a periodic low-RI SiO$_2$ (n ~ 1.5) nanodot array, fabricated by laser interference lithography on the anode, and contrasting the high RI organics. Optimized array parameters, evaluated via computational analysis, were 300 nm pitch, 60 nm height, and 0.3 coverage ratio. These parameters minimized spectral distortion and led to an increase of ~57.4% in the EQE of a green fluorescent device.

Feng et al [93] conducted one of the early studies on OLEDs on corrugated substrates. They fabricated 100 nm NPB/80 nm Alq$_3$/50 nm Ag OLEDs on 1-d and 2-d corrugated (depth ~ 70 nm, period ~ 550 nm) quartz/ITO substrates, and measured the significant EL through the nominally opaque Ag cathodes (figure 9). They attributed this EL to the coupling of the internal waveguide modes to the SPPs at the organic/Ag interface, the coupling of these to SPPs at the Ag/air interface with the assistance of the grating vector of the corrugation, and finally coupling of these to far field photons in the air beyond the Ag cathode.

In addressing SPP re-radiation, more recently, Liu et al [105] described OLEDs with 1-d patterned nanostructures based on soft nano-imprinting lithography. Specifically, they deposited a 3 nm MoO$_3$ hole injection and 70 nm NPB HTL on glass/ITO by standard thermal evaporation, and then patterned that NPB layer by soft imprinting by a PDMS mold. They tuned the corrugation period to 250 nm as their finite difference time domain (FDTD) simulations showed that at that period the SPP mode could re-radiate its energy. Following the corrugation step, they evaporated the 50 nm Alq$_3$, 1 nm LiF, and 80 nm Ag layers. The EQE of the resulting OLEDs exhibited a 20% enhancement relative to similar flat devices.
In another approach, Bi et al. [94] demonstrated outcoupling enhancement, attributed to broad band SPP mode outcoupling in WOLEDs based on two emitters, an orange phosphorescent and a blue fluorescent, with metal electrodes (one of which was a semi-transparent 15 nm Au and the other 80 nm Al) fabricated on a 2D grating of a dual-period corrugated structure. The periods were 225 nm and 325 nm with features’ depth of ~60 nm. The dual corrugated OLED showed satisfactory viewing angle characteristics. Experimental results and simulations showed a 48% increase in the EQE to 7.1% in comparison to planar devices.

Some substrates with buckling or periodic structures described next were also shown to enhance the outcoupling by reducing the internally waveguided light, as well as mitigating the SPP loss [106–110]. Buckling structures, however, can lead to shorts in the devices and some periodic structures lead to wavelength and viewing angle-dependent spectra. Overall, though, for SSL commercialization, it is currently preferred to further improve light extraction from the more familiar and well-studied planar devices devoid of extraction structures that can affect adversely the OLEDs’ optoelectronic properties.

Koo et al. [106] showed a random buckled structure formed spontaneously on preheated PDMS with a thin 10 nm Al film; the buckling occurred due to differences in the thermal expansion coefficient of the layers, as the PDMS/Al cools down to room temperature (figure 10). To increase the depth, the Al deposition and cooling cycles were repeated; the Fourier transform of the AFM images showed a quasi-periodic buckling structure. Green fluorescent Alq3 OLEDs were fabricated directly on the buckled structures, and their luminous and power efficiencies were enhanced by >2× across the entire visible spectrum, without introducing spectral changes or directionality.

Koo et al. [109] noted that for the above-mentioned buckled structure an imprinting process is required to transfer the pattern to the device substrate. To eliminate the imprinting and transfer processes, they fabricated the OLEDs directly on a hardened, buckled PDMS surface via a low-cost, scalable lithography- and imprinting-free process (figure 11) [111]. The buckled substrate’s peak (quasi)periodicity was ~0.6 μm and the depth was ~90 nm. It was fabricated similarly to the aforementioned structure, by thermal
evaporation of a thin 15 nm Al layer on PDMS and cooling that resulted in a buckled structure due to differences in the thermal expansion coefficients of the layers. UV exposure hardened the PDMS. The device shown has two green EMLs and it showed increased current and luminance in comparison to a reference device. The PE increased by 87% to 56.7 lm W\(^{-1}\) from 30.3 lm W\(^{-1}\) of a reference device.

In a follow-up to their first report [107], Koo et al [108] reported enhanced light extraction by defective silica sphere arrays with local hexagonal-closed-packed structure but no long-range order. PDMS replicas were first made using the silica sphere array substrate as a template. Then the PDMS replicas were stamped onto a UV-curable resin-coated glass substrate. The ITO/organic/LiF/Al OLEDs were fabricated on the glass/corrugated resin substrate, with the corrugation assumed to be maintained throughout the stack. The resulting fluorescent OLEDs exhibited broad band Lambertian emission, with the current and power efficiencies enhanced by 1.7\(\times\) and 1.9\(\times\), respectively, relative to reference devices. FDTD calculations showed that nanostructures on top of white emitters enhance light extraction with no wavelength dependency [112], as was also shown experimentally for some other patterned and planarized WOLEDs, as shown below, and when MLAs or scattering layers are added.

An EQE of nearly 55% at 1,000 nits was achieved by Ou et al [113] for corrugated OLEDs on internal ‘deterministic aperiodic nanostructures’ (‘DANs’) with long-range disorder. The devices were fabricated on glass/ITO/PEDOT:PSS by soft nanoimprinting lithography of the PEDOT:PSS. The structures also included an external DANs imprinted on resin deposited on the blank (air) side of the glass substrate, which was subsequently UV-cured.

Other types of quasi-periodic patterned OLEDs were also evaluated for their capability to enhance light outcoupling. Youn et al [114] demonstrated this effect for a corrugated OLED on sapphire (\(n \sim 1.8\)). Figure 12 shows the schematics of the device structure. The pattern was generated by first using the Langmuir–Blodgett technique to self-assemble arrays of silica spheres of 275 nm in diameter on the substrate. Inductively coupled plasma—reactive ion etching on one side of the sapphire generated the pattern and buffer oxide etching removed the remaining spheres on the opposite side. The pitch of the generated structure was \(\sim 260\) nm with \(\sim 50\) nm FWHM (indicating a broad range of grating vectors with all azimuthal directions covered). The height of the features was 80–90 nm. The use of a corrugated high \(n\) sapphire substrate together with an IMF and hemispherical lens resulted in an EQE of 63% at 225 cd A\(^{-1}\) for an encapsulated green OLED relative to \(\sim 50\%\) in a planar device.

In a recent study of OLEDs on a structured anode, Samigullina et al [115] fabricated and evaluated OLEDs on 2-d TiO\(_2\) block arrays, with a square cross section, to extract the internally waveguided light and mitigate SPPs. They varied the width of the block from 400 to 1,000 nm and the spacing between the blocks from 200 to 1,000 nm (i.e., the period varied from 600 to 2,000 nm) and fabricated bis(2-methyldibenzo[f,h]quinoxaline) (acetylacetonate) iridium(III) [Ir(MDQ)\(_2\)(acac)]-based OLEDs (i.e. with preferentially horizontal transition dipole moments) on the glass/ITO/TiO\(_2\) block arrays. Using a hemispherical lens to extract the substrate mode they obtained a maximal EQE of 45.2%.

Note that the above examples provide various structures, including patterns of different pitch and height values. A more systematic study regarding these parameters was performed recently using plastic substrates [116, 117].
3.2.4.2. Corrugated top-emitting OLEDs (TEOLEDs)

Several studies on corrugated TEOLEDs have been reported [118–121]. Such devices are attractive due to the ability to fabricate them directly on electronic circuits for e.g., active matrix displays. Jin et al [118] demonstrated that periodically corrugated Ag cathodes resolve the tradeoff between the lower stability of devices with thin Ag layers, which results from noncontiguous layers, pinholes, etc., and the lower transmission of thicker films. They compared 1-d and 2-d corrugated (350 nm period) Alq3 OLEDs with a top Ca/45 nm Ag cathode and a 40 nm NPB layer to flat devices with a top Ca/20 nm Ag cathode. They found that while the peaks of the EL spectra were angle-independent, their lineshape modestly red-shifted with viewing angle. Importantly, the luminous efficiency at ∼1,000 units of the corrugated devices was enhanced by ∼50% (from ∼4 to ∼6 cd A⁻¹), in spite of the relatively thick top Ca/Ag cathode of the corrugated devices. Lower or no enhancements have been reported by Schwab et al [119]. They described coherent mode coupling in phosphorescent red Ir(MDQ)₂(acac)-based TEOLEDs fabricated on 1-d corrugated substrates with periods of (a) 0.6 µm (corrugation height h = 144 nm) and (b) 1.0 µm (h = 69 nm). They reported that the ∼12% EQE of device (a) was actually lower than that of the planar device, which was ∼15%. The EQE of device (b) was 17.5%.

Xiang et al [120] fabricated flexible white transparent TEOLEDs with an impressive maximal EQE of >72%, PE approaching 170 lm W⁻¹, and a CRI > 84, with nanostructured composite electrodes. This study is described in additional detail in section 4 below, which surveys OLEDs on flexible substrates.

Kim et al [121] added a light-scattering dielectric layer to their high RI (n = 1.8) planarized TEOLED (figure 13). They observed that this outcoupling structure increased the EQE from 15 ± 2% to 37 ± 4%, a 2.5× enhancement over the reference device with a metal mirror. Importantly, there was no need for MLA or index matching layers, and simulations indicated that the enhancement can be 3.4× in comparison to conventional TEOLEDs that have two metal electrodes (one semi transparent), and hence a strong cavity effect.

In the structure shown in figure 13, light is emitted from the top or from the bottom. In top-emitting green and white OLEDs, SPP elimination was achieved by replacing the bottom metal electrode with ITO on a Teflon (90%–98% reflection in the visible; n ∼ 1.35) diffuse reflector planarized by a high n layer, where light is waveguided until it reaches the Teflon surface, where it exits into the viewing direction due to repeated scattering.

We note that in comparing the EQE of an OLED with a light-outcoupling structure to the control device, to determine the enhancement factor, one needs to be aware of cavity effects in the control device, and possibly in the devices with the outcoupling structures as well. This issue was recently treated in detail by Fu et al [95].

3.2.4.3. OLEDs on planarized patterns

Various planarized periodic and non-periodic sub-anode designs have been evaluated [121–123]. Besides the aforementioned study of the planarized TEOLED [121], bottom-emitting sub-anode extracting structures composed of low-RI SiO₂ nanoparticles embedded in a high-RI TiO₂ host matrix, both unplanarized and planarized with an additional PECVD (plasma-enhanced chemical vapor deposition) SiO₂ layer were
fabricated on glass [122]. The unplanarized and planarized structures resulted in an increase of ≈50% and ≈100%, respectively, in the outcoupled power and current efficiencies. In planarized phosphorescent green devices, they reached ≈115 lm W⁻¹ and ≈100 cd A⁻¹, respectively, at ≈2 mA cm⁻².

In another example, Qu et al [123] described a buried sub-anode grid. The lower n glass grid was filled (planarized) with high-\(n\) TiO₂ for outcoupling the internally waveguided light into the substrate. Another TiO₂ grid served to extract the substrate mode. In a green OLED, the EQE increased from ≈15% to ≈18% (i.e. by 20%), and then to ≈40% with the additional TiO₂ grid. The PE increased from 36 to 43 to 95 lm W⁻¹, respectively. The fabrication involved photolithography and wafer bonding.

In a promising approach, Kim et al [124] proposed a random scattering layer (RSL) for enhancing light outcoupling; figure 14 shows the structure used for calculations as well as experiment. The RSL consisted of glass or SiO₂ nanostructures with random period and size, which has the potential to overcome EL spectral shift and angular dependence seen with diffracting periodic scattering structures. The RSL was planarized with a high-index buffer layer; light that enters that layer is forward scattered by the RSL. Calculations
showed an increase of 37% in the EQE when compared to a reference OLED that provided the highest experimental EQE. Interestingly, but not surprisingly, they found that light absorption in the ITO layer is a very significant factor in determining the enhancement because the internally waveguided light is significantly attenuated by ITO absorption before it is extracted. For typical ITO the EQE of their WOLEDs increased from $\sim 23\%$ to 33%, i.e. a $\sim 43\%$ enhancement, but it could be increased by $\sim 80\%$ for a much lower extinction coefficient. They also noted that this absorption coefficient of ITO can be controlled to some degree by the deposition conditions [125, 126].

Other promising enhancing structures using high index planarizing layers have been evaluated [127–130]. Jeon et al [127, 128] described WOLEDs with an EQE > 50% and low roll-off that were fabricated by inserting a vacuum nanohole array (VaNHA) into PhOLEDs; the planarization layer was Si$_3$N$_4$. Figure 15 shows the schematics of the structure and the mode fraction/EQE vs the ETL thickness of the bare and VaNHA substrate with and without a hemispherical lens. By comparing the observed behavior with optical modeling results they concluded that the VaNHA extracts the entire internally waveguided light. Indeed, when combined with a hemispherical lens to extract the substrate mode, they obtained a maximal EQE of 78%, and 75.9% at 1000 cd m$^{-2}$. While these are the highest reported EQEs for WOLEDs, their fabrication is somewhat complex and potentially expensive. The EQE with the hemispherical lens but without the VaNHA was 37.1%, i.e. the latter resulted in a 2.1 $\times$ outcoupling enhancement.

Internal scattering layers were fabricated and evaluated by Cho et al [129]. Indium zinc oxide (IZO)/organic/Al OLEDs were fabricated on glass/organic wrinkles/high index planarization layer as the internal scattering structures. The solution-processed wrinkle structures, fabricated by exposure of a spin-coated liquid prepolymer layer to UV, were planarized with a high-index Pixelligent layer based on ZrO$_2$ nanoparticles. The resulting device exhibited both reduced surface roughness and optical haze. The maximal EQE was 24.2%, or 1.24 $\times$ the control device with no internal scattering layer. When adding a MLA or a hemispherical lens to the air-side of the glass the maximal EQE reached 30.5% (1.56 $\times$) or 41.8% (2.14 $\times$), respectively.
In a promising approach, Qu et al [130] described a sub-electrode hemispherical microlens array (SEMLA), shown schematically in figure 16. The SEMLA was etched in glass and the pattern was planarized with a high-index spacer layer \( n \sim 1.8 \) on which the OLED was fabricated. An IMF at the glass/air interface was employed to couple to a Si photodiode for maximal outcoupling. The maximal EQE for a green OLED was \( \sim 70\% \) and for a WOLED \( \sim 50\% \) at 100 cd m\(^{-2}\), with the SEMLA strongly reducing the internally waveguided loss.

In an earlier conceptually similar approach, Koh et al [98] planarized photolithographically patterned (6 µm pitch) 150 nm thick high-index ITO \( (n_{\text{ITO}} \sim 2) \) with PEDOT:PSS \( (n \sim 1.42) \). Due to the strong contrast between these RIs, internal reflections and scattering at the ITO/PEDOT:PSS interfaces, together with an added MLA, resulted in a \( \sim 52\% \) enhancement in the light outcoupling from a planar fluorescent OLED relative to a standard device without the structure or the MLA. The authors noted that the emission was largely wavelength and viewing angle independent.

We note that OLEDs on a high index layer that planarizes buried patterns in plastic substrates, which are easily fabricated by direct imprinting methods, result in similar large EQE enhancements [131]. Flexible OLEDs are surveyed below.

### 4. OLEDs on flexible substrates

Flexible OLEDs are of great interest for applications, such as displays, SSL, and wearable printed sensors [5, 116, 132], including in some emerging medical applications [133, 134]. Thin glass (e.g. Willow glass, Corning) is often the preferred flexible substrate due to its transparency and very low water vapor and oxygen permeation, though its handling is not simple. Flexible displays are currently produced by e.g., Samsung and LG, and flexible SSL panels by LG and Konica Minolta [5] as well as by OLEDWorks LLC. Devices on thin plastic substrates are advantageous for e.g., medical applications due to their lighter, stretchable, bendable, and foldable properties.

Plastic, is, of course, the most common type of flexible substrate material studied for OLEDs. The disadvantage of plastic is its unacceptable permeability to water vapor and oxygen, and in some materials a reduced thermal stability. Development of efficient water and oxygen barrier films will advance the use of plastic substrates. Examples of studied various plastic substrates for different applications include polyethersulfone (PES) [135], polycarbonate (PC) [116, 136], polyethylene terephthalate (PET) [116, 137–141], polyimide (PI) [55, 142–145]) (colorless PI, \( n = 1.63 \)), and the \( n = 1.56 \) Norland optical adhesive NOA63 [146].

The selected examples summarized below include studies of plastic substrates with embedded scattering particles, OLEDs with a PEDOT:PSS anode on plastic, and flexible OLEDs on planar, patterned, and planarized buried structures in plastic.

Shen et al [145] studied 5–25 µm thick colorless PI films doped with light scattering 200 nm Al\(_2\)O\(_3\), 200–300 nm SiO\(_2\), or 400–600 nm SiO\(_2\) particles for enhanced OLED light extraction. They evaluated the doped PI characteristics, such as scattering efficiency, haze, and transmittance, including the effect of the RI of the particles relative to that of the PI, the particle size, and the doping level. However, they did not provide results on OLEDs with these scattering films.
Another multifaceted study combining flexible plastic substrates/electrodes with light-scattering particles, which showed the promise of this approach for enhancing OLED light extraction in e.g., lighting applications, was reported by Tong et al [147]. Substrates with silver nanowire (AgNW) embedded within an acrylate ($n \approx 1.53$) mixed with $\sim 200$ nm high-index barium strontium titanate nanoparticles ($n \sim 2.5$) and PEDOT:PSS HIL resulted in WOLEDs with a PE of $107 \text{ lm W}^{-1}$ and EQE of nearly 49% at 1,000 cd m$^{-2}$. This was an enhancement of $\sim 2.75 \times$ in the EQE of an optimized device relative to a device on glass/ITO (EQE $\sim 18\%$). The EQE of a reference device on the AgNWs/acrylic matrix was $\sim 23.5\%$.

There are various reports on other flexible substrates with OLEDs comprising ITO-free electrodes. As an example, Liu et al [148] fabricated enhanced-efficiency Alq$_3$-based OLEDs on smooth, treated PEDOT:PSS anodes. The smooth electrode was generated via a template stripping process. PEDOT:PSS was first spin-coated on glass followed by spin-coating a $\sim 400 \mu$m thick photopolymer on it. Following UV curing the photopolymer/PEDOT:PSS was stripped off the glass, exposing the smooth PEDOT:PSS surface to which the authors attribute the $\sim 33\%$ efficiency enhancement, from a maximum of $\sim 4.5$ to $\sim 6 \text{ cd A}^{-1}$.

Replacement of ITO by PEDOT:PSS in flexible OLEDs has also been reported in other studies [116, 149]. While ITO is of high transmittance and conductivity, it is not ideal for plastic substrates, as it is brittle and requires relatively high-temperature deposition that is not compatible with most plastic materials. The addition of AgNWs or other electrode designs have been proposed to improve flexible OLEDs with ITO-free electrodes [150]. Another alternative to ITO is dielectric/Ag/dielectric electrodes [5, 151–159].

Enhanced efficiency was also observed in a flexible green PhOLED fabricated on a conductive, colorless PI substrate embedded with AgNWs and smoothed by a PEDOT:PSS layer. The enhancement was partially due to scattering by the AgNWs and some extraction of the waveguided light [55].

Various experimental and theoretical studies of patterned structures were reported [116, 145, 146, 160–164] indicating reduced internal waveguiding and SPP losses. The outcoupling enhancement is typically attributed to diffraction and scattering in the periodic patterns [4, 34, 95, 165], and to avoid directional EL, quasi random nanopatterns were suggested [138, 140]. MLAs have the same effect on the angular profile, resulting in a nearly Lambertian EL.

Efficient WOLEDs, reported by Li et al [166], were fabricated on quasi-periodic nanostructures with broad distributions of periodicity and depth that were produced controllably by reactive ion etching of PDMS. The nanostructures were applied to two-unit tandem WOLEDs and together with a hemispherical lens the EQE reached 76.3% by extracting the internally waveguided and the substrate modes, as well as minimizing SPP loss; the EL showed no angular- or wavelength-dependence. The authors state that as the nanostructures were generated directly in PDMS, they are compatible with emerging flexible devices.

Integrated substrates, which include both nano- and micro-patterns, can be easily fabricated on flexible substrates. Such structures were integrated also using glass and other substrates as reviewed by Feng et al [3], though in the latter the microstructures were attached to the air-side of the substrates.

The overarching advantage of flexible substrates, particularly plastic, is their potential to enable a R2R manufacturing process, which can lower the manufacturing costs drastically. For example, R2R produced hot stamp foils for holographic security films costs $0.10$ to $0.30 \text{ m}^{-2}$ [167]. We therefore next elaborate on a potential R2R process for OLED substrate fabrication.

The R2R fabrication of patterned substrates for enhancing light outcoupling is attractive also due to the potential low cost versatile fabrication of a variety of periodic and aperiodic designs in different materials and, importantly, with the designs transferable to glass. The patterns can be planarized and include, in addition to a top nanopattern (on which the OLED is fabricated), the micro-patterned MLA at the air side in the same substrate. Moreover, all of these provide an easy-to-use tool for advancing the knowledge in the field of light extraction. Various lithographic, imprinting, and transfer techniques and as mentioned, various materials can be used for forming patterns. For example, the photopolymer NOA63, mentioned above, was used as both the flexible substrate and a patterned encapsulating film for Alq$_3$ TEOLEDs with an opaque Ag anode and a thin (semitransparent) Al/Ag cathode [146].

Figure 17 illustrates a cost-effective R2R approach by MicroContinuum Inc. (www.microcontinuum.com), of imprinting a PC substrate with a top nano-array and a bottom micron-size MLA to generate an integrated substrate that will also include the anode and a metal mesh for enhanced conductivity [3].

Beyond the foregoing process, MicroContinuum Inc. directly imprints patterns in substrates, such as PC (figure 17) and additionally employs two-layer formulations in substrates, such as PET/thermoplastic ester. Each technique clearly has its own advantages [116] enabling high throughput.

As mentioned, notwithstanding the strong potential advantage of a R2R manufacturing process, one of the main issues with plastic substrates is their relatively high permeability to water and oxygen, which adversely affect the OLEDs’ lifetime and hence long-term performance. Mechanical, thermal and chemical instability are also potential issues in some cases [168–170]. Various encapsulation approaches have been...
developed, including hybrid organic/inorganic multilayer coatings [168–171]. However, approaches that are commercially more adequate are flexible laminated thin films [172]. Suitable barrier films that are yet to become widely available may help mitigate this issue.

Zhou et al. [163] fabricated flexible green and white PhOLEDs on a plastic substrate as shown in figure 18. A template of glass/PDMS/10 nm Al was first used. It was heated to 100 °C and then cooled to generate a buckling of variable periodicity and 50–70 nm depth. The pattern was transferred to a resin on glass that was UV cured and ozone treated. For device fabrication, an AgNW mesh was embedded in a nano-imprinted PET substrate on which PEDOT:PSS was deposited. A MLA was added at the air-side of the PET. For the green PhOLED the EQE and PE increased 2.6 × to 50% and 106 lm W⁻¹, respectively, at 1,000 cd m⁻² [163]. The design was complex and included a Ni mold photolithography and pattern transfer as well as silver paste scraping. The transmittance was >87% and the design was dependent on time and pressure during the imprinting.

Figure 19 shows AFM images of convex and concave patterns in PC and PET/CAB (cellulose acetate butyrate), respectively, which showed enhanced outcoupling from OLEDs to various degrees, depending on the period (pitch) and height of the features, as well as the uniformity of the substrate [116]. The structures were fabricated using the R2R nanoimprinting facility of MicroContinuum Inc.

For example, a green PhOLED of the structure PC-320/PEDOT:PSS/MoOₓ (1 nm)/10% MoOₓ:NPB (22.5 nm)/NPB (22.5 nm)/6% Ir(ppy)₂:CBP (11 nm)/Bphen (40 nm)/LiF (1 nm)/Al (100 nm) on PC, with a pitch of ~750 nm and features’ height of 320 nm (hence the notation PC-320), showed EQEs of 36%–50% (see below). The EQE on flat glass/ITO was 18% and on flat PC/PEDOT:PSS 22%. The enhancement was shown to be due to diffraction and hence mitigation of SPPs and reduction of internal waveguided light [116, 165]. Figure 20 shows the angular dependence of the EL in comparison to a Lambertian distribution. Some structure is seen in the distribution, which is relatively broader than expected, though later experiments indicated that the patterned substrate may act partially as a MLA, which alleviates the directionality expected due to diffraction. In addition, the EML, which is also undulated, results in transition dipole moments with random orientation relative to the undulating plane of the device. This should also result in an essentially
Figure 19. AFM images of corrugated PC (left) and PET/CAB (right). The full scale is 10 µm. Reproduced from [116] with permission from John Wiley & Sons.

Figure 20. (a) The angular dependence of the EL for a green PhOLED on PC-320 with a ∼750 nm pitch. (b) EL spectrum of devices on flat and patterned PC substrates with a ∼750 nm pitch and various heights. Reproduced from [116] with permission from John Wiley & Sons.

Lambertian profile. The figure also shows the EL spectrum for PC substrates with a ∼750 nm pitch and heights h varying from ∼215 to ∼500 nm. As seen, there are small variations, mostly for larger h values.

Other device structures provided larger efficiencies, though the enhancement was typically 1.6× to 2× relative to a reference planar device on plastic [152–158]. The PEDOT:PSS however, is not as conductive as ITO and often, as in the examples above, AgNWs were used together with it.

Figure 21 [116] shows results on green Ir(ppy)$_3$-based PhOLEDs with the structure: Anode/HAT-CN (5 nm)/10% MoO$_3$:TAPC (120 nm)/TAPC (20 nm)/6% Ir(ppy)$_3$:mCP (20 nm)/TmPyPh (20 nm)/20% CsF:TmPyPh (40 nm)/LiF (1 nm)/Al (100 nm), where the substrate/anode was glass/ITO, glass/PEDOT:PSS, flat PC/PEDOT:PSS, and PC-320/PEDOT:PSS; PC-320 is the patterned substrate with pitch ∼750 nm and h ∼ 320 nm.

The patterned devices’ maximal EQE was ∼50%, a 2× enhancement relative to the planar device on PC/PEDOT:PSS.

As mentioned, plastic substrates require barrier films to prevent oxygen and moisture penetration. Hence, generation of polyester corrugations transferred to glass substrates may currently be a better approach for enhancing light outcoupling. Another challenge observed with patterned substrates is the change in the pattern as the OLED layers are formed. That is, experiments [131] indicated a decrease in the pattern features’ height with added layers. This is seen particularly in patterns of small pitch and height as seen in figure 22. In contrast, as expected, the larger the pitch, the more conformal the OLED layers are. Additionally, solution processed layers lead to the largest change in the pattern height with thicker layers in the troughs relative to the top and sides of the features. Thermal evaporation with broad linear sources and moving substrates can overcome this non-uniform layer deposition.

We note that the initial increase of the EQE vs brightness (at low current densities) for OLEDs on plastic substrates and, particularly when patterned, is pixel-dependent and hence is likely due to burning of shorts (that eliminates them) possibly associated with defects or impurities. This situation is accentuated as the measurements are performed on unencapsulated devices.
Figure 21. The EQE vs brightness of green PhOLEDs on different substrate/anode pairs. The devices were not encapsulated, likely causing the initial rapid rise in the EQE of the corrugated device PC-320 at low brightness as shorts due to defects are burned and eliminated. Reproduced from [116] with permission from John Wiley & Sons.

Figure 22. Focused ion beam (FIB)—SEM images of non-conformal green OLEDs on convex PC substrates [131].

Figure 23. EQE vs brightness for a green PhOLED on glass and on a planarized buried structure, where the planarization layer was of high RI. The anode was ITO. Extracting the substrate mode resulted in an EQE of ∼60%. The schematic of the structure (not to scale) is also shown [117]. As with planarized buried patterns in glass, OLEDs on planarized buried patterns in plastic, with the planarization layer being of high RI, show significant enhancement in the EQE [117]. Figure 23 shows an example of the enhancement prior to extracting the substrate mode with an IMF, which then leads to a maximal EQE of ∼60% depending on the structure of the planarized buried structure. The figure also shows a schematic of the design.

Finally, as mentioned in section 3.2.4.2. on corrugated TEOLEDs, Xiang et al [120] fabricated flexible white transparent TEOLEDs with an impressive maximal EQE of >72%, PE approaching 170 lm W⁻¹ and a CRI > 84, with nanostructured composite electrodes. To improve anode conductivity, the OLEDs were fabricated on a hexagonal (100 µm period) Ag grid embedded on the PET substrate. An 80 nm thick PEDOT:PSS layer was then spin-coated onto the Ag grid-PET, following which an internal moth-eye pattern (period ∼ 350 nm) was imprinted with a PDMS mold. Next, three phosphorescent layers were deposited on
the patterned PEDOT:PSS. Finally, the top transparent electrode of LiF/ultrathin Al seeding layer/Ag layer/NPB capping layer completed the device.

5. Summary and concluding remarks

In summary, we presented advances and challenges of light extraction from OLEDs. Light outcoupling from conventional bottom-emitting OLEDs is typically \( \sim 20\% \), due largely to losses to the external, substrate waveguide mode, the internal waveguide mode (between the metal cathode and the anode/substrate interface) and surface plasmon-polaritons (SPPs) at the metal cathode/organic interface. We reviewed some of the numerous studies that focused on increasing the outcoupling factor \( \eta_{\text{out}} \) by addressing one or several of the loss mechanisms in combination. These included (a) extracting the substrate mode via use of MLAs, hemispherical lens, scattering layers, and IMFs, (b) enabling all generated photons to reach the substrate, i.e. minimizing internal waveguiding and SPPs, by fabricating ITO-free OLEDs, using low-RI organic layers and/or high-RI substrates, (c) reducing internal waveguiding and SPPs by using emitting molecules with preferential horizontal transition dipole moments, and (d) outcoupling light employing enhancing structures. Among the latter, we reviewed OLEDs on PC or PC-inspired structures, as well as scattering layers. Special attention was given to bottom- and top-emitting OLEDs on corrugated substrates (i.e., patterned OLEDs), and OLEDs on substrates with buried patterns (i.e. planarized substrates). Moreover, we reviewed various studies of flexible OLEDs fabricated on plastic, which provide a tool for facile fabrication of extraction enhancing substrates. Flexible organic electronic devices are gaining interest in biomedical applications. As in displays, there is also interest in flexible devices on thin glass, including in automotive applications where light extraction remains a challenge.

Other remaining challenges include the actual extraction of essentially all of the external waveguided light, given the observation that all of the laboratory studies that attained high \( \text{EQE}_{\text{max}} \) utilized a large hemispherical lens and/or IMF, which are incompatible with industrial applications. Another remaining challenge is the ease of fabrication of enhancing structures and hence their cost and throughput, in particular for upscaling. Such structures should not be too complex, as some are. They should preferably be compatible with planar OLEDs to avoid spectral and viewing angle dependence of the EL. Hence, despite significant advances, R&D efforts are still ongoing.

Data availability statement

No new data were created or analysed in this study.

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ORCID iD

Ruth Shinar https://orcid.org/0000-0001-9936-7723

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