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REVIEW

Recent advances in light outcoupling from white organic light-emitting diodes

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Abstract. Organic light-emitting diodes (OLEDs) have been successfully introduced to the smartphone display market and have geared up to become contenders for applications in general illumination where they promise to combine efficient generation of white light with excellent color quality, glare-free illumination, and highly attractive designs. Device efficiency is the key requirement for such white OLEDs, not only from a sustainability perspective, but also because at the high brightness required for general illumination, losses lead to heating and may, thus, cause rapid device degradation. The efficiency of white OLEDs increased tremendously over the past two decades, and internal charge-to-photon conversion can now be achieved at ~100% yield. However, the extraction of photons remains rather inefficient (typically <30%). Here, we provide an introduction to the underlying physics of outcoupling in white OLEDs and review recent progress toward making light extraction more efficient. We describe how structures that scatter, refract, or diffract light can be attached to the outside of white OLEDs (external outcoupling) or can be integrated close to the active layers of the device (internal outcoupling). Moreover, the prospects of using top-emitting metal–metal microcavity designs for white OLEDs and of tuning the average orientation of the emissive molecules within the OLED are discussed. [DOI: 10.1117/1.JPE.5.057607]

Keywords: organic light-emitting diodes; white light; solid-state lighting; outcoupling; emitter dipole orientation; thin-film optics.

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1 Introduction

The idea of using organic compounds with extended conjugated electron systems for efficient, sustainable, and flexible generation of light has fascinated scientists around the world for nearly three decades now. Within this timeframe, the efficiency, brightness, and stability of organic light-emitting diodes (OLEDs) have improved dramatically. Although OLEDs were initially developed mostly as the active element in emissive displays, the technology has matured to a level that now brings within reach applications in general lighting, a field that poses in many regards much stricter requirements on device performance. General lighting requires OLEDs to generate balanced white emission, produce high brightness levels, and to do so at competitive efficiency when compared to established technology, in particular, fluorescent lamps and conventional LEDs. For many years, a primary focus has been to improve the internal efficiency of OLEDs, i.e., the yield of charge-to-photon conversion. However, in many state-of-the-art devices, this is now achieved with near unity efficiency, mostly thanks to the development of triplet harvesting,[1–3] doped charge transport layers,[4,5] and efficient charge blocking structures.[6] Over the past years, the focus has, therefore, moved toward improving the efficiency of outcoupling, i.e., the fraction of the generated photons that can be extracted from the device into the

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surrounding air. Despite some good progress, outcoupling remains a major loss channel with well <30% of the photons being extracted from typical OLEDs. Developing efficient and practical concepts for light extraction, in particular for white-emitting OLEDs, thus remains a major challenge in the field.

In this article, we review the recent progress in the field of white OLEDs, with particular emphasis on recent developments for improved outcoupling. Several other review papers have been published over the past few years that cover other aspects of white-emitting OLEDs.[7–12] Following a brief description of the main concepts used for generating white emission from organic materials, we will summarize the processes leading to limited outcoupling efficiency in OLEDs. We then discuss a series of outcoupling concepts and give examples of their implementation in white OLEDs, starting from the more established external outcoupling structures where a light extracting microstructure is applied externally to the OLED. We then cover internal approaches, i.e., approaches employing structures that are located within the optical near-field of the device's active area. We will also discuss the prospect of white top-emitting OLEDs and work on tuning the spatial orientation of the light-emitting molecules within the device to further enhance light extraction efficiency.

2 OLEDs and Approaches to Generation of White Light

The general principle of operation of OLEDs has been described in various review papers and book chapters (see, e.g., Refs. 13 and 14). Most state-of-the-art OLEDs today are multilayer devices in which the light-emitting materials are sandwiched between a series of other organic semiconductors to optimize the injection of charge, improve device stability, and tune the device thickness. In particular, the use of molecularly doped charge transport layers has proven highly useful as it allows circumventing charge injection barriers and thus facilitates driving voltages close to the energy of the emitted photons.[4,15] These layers are then embedded between two electrodes, a cathode and an anode, at least one of which is (semi)transparent. It is useful to distinguish between bottom- and top-emitting devices: in the former, light is emitted through the bottom electrode and the substrate of the device, whereas in the latter, the top contact of the OLED facilitates emission of light. Therefore, bottom-emitting devices obviously require a transparent substrate—typically glass or transparent plastic films—while top-emitting structures can be used on opaque substrates, e.g., metal foil or silicon chips containing driver electronics. However, all OLEDs reported to date require encapsulation to prevent degradation by ambient oxygen and humidity; in the case of top-emitting OLEDs, the encapsulation has to be optically transparent.

OLEDs are also frequently classified according to the method used for their fabrication and whether they are based on monomeric compounds (small molecules) or higher molecular weight polymers. Traditionally, small molecule OLEDs have been fabricated by subsequent deposition of the different materials onto a substrate using thermal evaporation under high vacuum. By contrast, polymer OLEDs are generally obtained by solution-based approaches, predominantly spin-coating or printing. Nowadays, there are various hybrid approaches intended to harness both the ease of solution-based processing and the superior efficiency and stability provided by small molecule devices.

Compared to the emission from conventional inorganic semiconductors, organic materials generally tend to show broad emission spectra, with the full width half maximum of the spectrum typically in the range of 50 to 100 nm. There has been some work on broadening the emission spectrum of organic emitters even further to cover the entire visible part of the electromagnetic spectrum with a single white light-emitting material.[16,17] However, for the most part, the community has worked on combining the emission from several (typically two or three) different types of emitters, each generating a specific color, to cover the required range of the spectrum. In principle, there are several ways of combining the emission from the different emitters as described in some detail in Refs. 7 and 8. In brief, these approaches include stacking separate layers or even separate devices generating the different colors on top of each other. Alternatively, the different lumophores can be present within a single layer, which is achieved either by blending or by synthesis of a macromolecular structure—most often a polymer—that combines the different lumophores within one material. There is also a side-by-side approach in which OLEDs

emitting light of different colors are deposited next to each other[18,19] and a color-conversion strategy in which a blue-emitting OLED is partially covered with color-converting dyes or phosphors to achieve white emission.[20–22] These latter two concepts resemble structures widely used in inorganic white-emitting LEDs, but so far they have had limited success in the OLED community (for a number of technical reasons). In this review, we will, therefore, focus on devices based on the aforementioned stacking and blending approaches.

In early OLEDs, light was emitted by relatively simple fluorescent compounds that were often derived from commonly used laser dyes. While these achieve high fluorescence quantum yields, it soon became clear that during electrical operation, 75% of the excitons formed on the molecules have a triplet character and thus do not relax radiatively. To avoid this major loss channel, phosphorescent emitters, in which triplet states efficiently contribute to the generation of light, have been developed.[2] These emitters are typically heavy metal ligand complexes, frequently with iridium, platinum, or palladium cores. Recently, high efficiencies have also been obtained with copper-based emitters, which promise to be less expensive due to the absence of a rare metal core.[23,24] Today, most white OLEDs contain phosphorescent emitters, at least for the generation of the red and green components of the spectrum. Blue phosphorescent emitters with high efficiency and stability remain scarce, thus some white OLEDs employ a triplet harvesting approach, in which singlet and triplet excitons are initially formed on the same material, but triplets are then funneled to phosphorescent red or green emitters while singlets emit from a fluorescent blue emitter.[25–27] Very recently, carefully engineered metal-free emitter molecules with near-resonant singlet and triplet state energies have been demonstrated to recycle triplet excitons with near unity efficiency by a process called thermally activated delayed fluorescence (TADF).[3] TADF emitters covering most of the visible spectrum, including fairly saturated blues, are now available,[28] and there are first reports on the use of TADF emitters in white-emitting OLEDs.[29]

3 Outcoupling in Conventional Devices

Before moving on to discuss the various approaches to enhance outcoupling, it is useful to revisit how light is extracted from conventional planar OLEDs that do not contain any specific outcoupling structures. For a more complete description of this process, see Refs. 30–32.

In a highly simplified picture, the losses associated with light outcoupling can be attributed to reflection of the emitted light at the various interfaces inside the device followed by reabsorption and thermalization [Fig. 1(a)]. In a simple ray-optics picture and assuming that light is initially emitted isotropically within the emissive layer (EML) of the device, one can estimate the efficiency of light extraction as $\eta_{\text{out}} \approx 1/(2n^2)$, where n is the refractive index of the material in which the light is generated.[33] However, the $1/2n^2$ approximation is now generally considered to be an oversimplification and most modern models instead describe the emitting molecules/polymers in the device as classical oscillating dipoles and then apply a transfer matrix procedure to compute their far-field emission profile after interaction with all optical interfaces in the device. In this way, thin-film interference effects are properly described, which is important because they often cause substantial modulations in light extraction efficiency, e.g., when changing the distance between the emissive dipoles and the reflective metal electrode.

In order to properly model coupling of the emissive dipoles to the guided light modes of the structure (i.e., to account for all light that is lost by total internal reflection at the different interfaces), the dipole emission is usually developed as a function of the normalized in-plane wavevector k_x/k, rather than as a function of the external angle of emission [see Fig. 1(a)]. Here, k is the modulus of the wavevector within the EML, so that $k_x/k = 1$ would correspond to the in-plane propagation of light in a bulk EML. In addition to coupling to waveguided modes, dipoles can also couple their energy to electron plasma oscillations at the surface of the metallic electrode (s) of the device. These are typically referred to as surface plasmon polariton modes or evanescent modes. Plasmon modes are associated with in-plane wavevectors of $k_x/k > 1$ and a substantial proportion of the total energy is funneled into plasmon modes if the EML is located close to a metal layer (i.e., distance $\ll$ wavelength of light).

To complicate matters further, one finally also needs to consider that the presence of the various different modes to which dipoles can couple their energy (emission into the surrounding

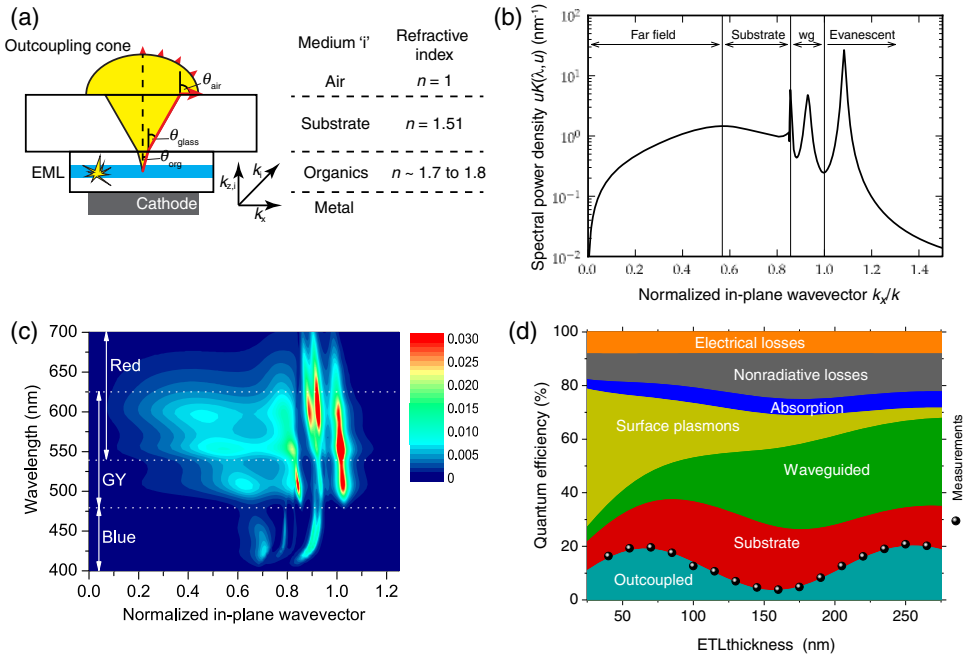

Fig. 1 Principles of outcoupling in organic light-emitting diodes (OLEDs). (a) Illustration of refraction of the light emitted in the emissive layer (EML) of a bottom-emitting OLED. The outcoupling cone corresponds to the range of emission angles within the EML for which light can be extracted and is not trapped by total internal reflection. (b) Power dissipation spectrum of a red bottom-emitting OLED for $\lambda = 610$ nm as a function of the normalized in-plane vector k_x/k.[30] (c) Two-dimensional power dissipation spectrum showing the spectral power density (false color) as a function of in-plane wavevector and wavelength for a four color white tandem OLED.[34] (d) Distribution of the different outcoupling loss channels in a red bottom-emitting OLED as a function of electron transport layer thickness as predicted by optical modeling. Experimental external quantum efficiency data for same device stack (circles).[35] [Reproduced with permission, courtesy of American Physical Society (b), AIP Publishing (c, d).]

medium, waveguided within the substrate or within the organic materials, plasmonic modes) affects the excited state lifetime of the emitting molecules and their probability to couple to each of the different modes. This phenomenon is known as the Purcell effect and needs to be taken into account to properly estimate the outcoupling efficiency of an OLED. In the model, this is achieved by introducing a coupling term to the differential equation describing the classical dipole.

It is worthwhile to note that for some time there was an aspiration of directly exploiting the Purcell effect to increase the overall efficiency of OLEDs. However, it is now clear that this is only possible for materials with low photoluminescence quantum yields where reducing the radiative lifetime means nonradiative relaxation of the molecules is out-competed, thus increasing their effective photoluminescence quantum yields. The emitters used in most of today's OLEDs show intrinsic photoluminescence quantum yields >70%, so the achievable efficiency gain is rather limited, in particular given that reducing the radiative lifetime typically requires coupling a considerable fraction of the dipole emission to plasmon modes, which do not contribute to the useful emission of the device.

Taking the above effects into account, a useful way of describing the outcoupling of light in a planar OLED is to consider the amount of power they dissipate in each direction or—using the wavevector picture—at different k_x/k. An example of such a power dissipation spectrum is shown in Fig. 1(b). The power dissipated in the range of $k_x/k < k_{\mathrm{air}}$ corresponds to the useful emission of the device; larger k_x/k corresponds to emission that is trapped within the substrate ($k_{\mathrm{air}} < k_x/k < k_{\mathrm{subst}}$), the active layers of the device ($k_{\mathrm{subst}} < k_x/k < k_{\mathrm{org}}$), or as plasmon modes ($k_x/k > k_{\mathrm{org}}$). Integrating over the different regions of the power dissipation spectrum and normalizing to the overall integral yields the fraction of energy coupled into each channel, with the integral of $k_x/k < k_{\mathrm{air}}$ giving the outcoupling efficiency.

In general, the power dissipation spectrum is wavelength dependent and thus needs to be computed over the entire emission range of the active material and then weighted with the emission spectrum of the respective emitter. For white OLEDs, the situation is complicated further by the fact that the emissive molecules generating the different spectral components are often located at different positions. Figure 1(c) shows the dissipation spectrum for a state-of-the-art tandem white OLED, illustrating how the different emissive layers each couple to the waveguide and plasmon loss channels.[34] By integrating over the complete spectral range and over appropriate intervals in k_x/k, one can then obtain a power budget, i.e., the fraction of power that is coupled to each of the different channels.

Several commercial and academic software packages have been developed that perform the above computations.[30,36–38] These also enable systematic variation of device parameters (e.g., the thickness of the different layers, choice of metals for electrodes, etc.) to optimize the outcoupling efficiency prior to the fabrication of actual devices. Figure 1(d) illustrates this as one of the most important parameters—the separation between the EML and the metal contact. However, as small variations in thickness and the refractive indices of the individual layers forming the device can strongly impact on device efficiency and because these parameters are difficult to control with sufficient accuracy in an experiment, empirical device optimization is still necessary in many cases to achieve the best possible performance.

4 External Outcoupling Schemes

A convenient way to categorize the different approaches of enhancing outcoupling is by the component of the trapped light that they extract from the device. External outcoupling structures are structures that are applied onto the backside of the substrate on which the OLED is deposited. These can generally be applied rather easily and without risk of negatively affecting the electrical operation of the OLED. However, such structures will only interact with light that is trapped in the substrate of the device, which somewhat limits the amount by which they can enhance device efficiency.

To prevent total internal reflection at the substrate—air interface of the device, the entire backside of the structure would ideally be curved with the active area of the OLED located at the center of a hemisphere. While such structures are widely applied for inorganic LEDs, which are near ideal point sources, they are generally considered impractical for large OLED lighting panels—one can hardly imagine enclosing a large OLED tile inside an even larger glass or plastic hemisphere. However, there have been several proof-of-principle demonstrations illustrating that the application of a hemispherical macroextractor can indeed increase outcoupling efficiency, by a factor of two easily for conventional device layouts (standard glass, no other optical enhancements).[39,40] Truncated prisms represent a slightly more compact alternative,[41] but ultimately also suffer from being too bulky when applied to large area devices. As a miniaturized version of these macroextractors, microlens and micropyramid arrays have been applied to the backside of OLED substrates.[42] If arranged in closed packing, such structures can cover a large fraction of the substrate, and, although less efficient than the bulky macroextractors, improvements in outcoupling efficiency by factors ranging from 1.5 to 1.7 have been reported.[22,42–45] In the micro-array configuration, some of the light hitting the surface of the lenses or prisms will be reflected back into the device (e.g., because the angle of incidence is above the critical angle of total internal reflection). However, by using OLED stacks with high reflectivity and low reabsorption, this light can be reflected onto the outcoupling structure again and again, until eventually the angle and location of incidence are such that the light can escape from the substrate. The efficiency of this photon recycling process is limited by the reflectivity of the OLED stack—in particular, by the reflectivity of the back metal contact. Photon recycling of light trapped in the OLED substrate does not necessarily require a well-defined optical microstructure. Instead, it is sufficient to have a structure that randomly and repeatedly alters the angle under which the trapped light propagates so that most of the light will eventually assume an angle within the outcoupling cone. Consequently, depositing a scattering film on the back-surface of the substrate or roughening the back-surface itself also increases the outcoupling efficiency.[46,47] A similar effect can be realized through a more complex conditioning of the glass/air interface.

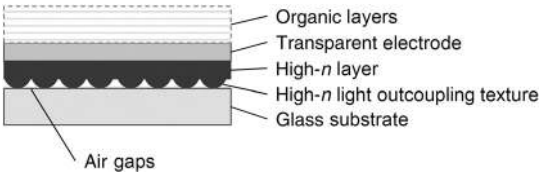

Fig. 2 Illustration of a light outcoupling structure used by Panasonic to fabricate white OLEDs with over 100 lm/W luminous efficacy. Rather than using expensive high refractive index substrates, a high-index thin film and outcoupling structure are combined with conventional glass substrates. (Reproduced with permission, courtesy of the Society of Photopolymer Science and Technology.)[51]

Yamasaki et al. proposed the application of ordered monolayers of silica microspheres on the glass surface to achieve enhanced scattering.[48] Cheng et al. fabricated microscopic mesh structures from poly(dimethyl siloxane) (PDMS), which they applied to the glass surface.[49]

It should be noted that the discussion above exclusively considers OLEDs fabricated on standard glass substrates (i.e., substrates with a refractive index around $n = 1.51$). Thus, between the organic materials (including the indium tin oxide (ITO) electrode) and the glass substrate, there is another step in the refractive index that confines many modes to within the organic layers. Switching to index matched substrates alleviates this effect, and external outcoupling schemes can then access the combined substrate/organics modes.[39] Under these conditions, a 2.4-fold improvement in outcoupling efficiency is possible. An even higher outcoupling efficiency (3.4-fold, when using a hemisphere) can be realized if one further suppresses coupling to metal surface plasmons, e.g., by designing devices to operate in the second optical maximum and using a thick spacer layer between the emission layer and cathode.[39] The drawback of refractive index matched substrates is clearly cost, and there is currently no direct pathway to bringing down the prize of high refractive index substrates to a degree that would facilitate upscaling to high-volume production. The use of a double-layer substrate has recently been demonstrated by a team at Panasonic to be a viable alternative to index matched glass substrates.[50] In this approach, a high-index polymer film [e.g., polyethylene naphthalate (PEN), $n = 1.77$] comprising a microlens array (MLA) is laminated with a conventional glass substrate so that the lenses face the glass substrate and local air gaps are formed (Fig. 2). By depositing a proprietary white OLED stack (phosphorescent, tandem architecture) on top of this structure and applying thin-film antireflection coatings to both sides of the glass substrate, a luminous efficacy of 133 lm/W and an external quantum efficiency (EQE) of 56% per tandem unit have recently been achieved.[51]

In recent work by Yoo et al., the high-index PEN was used as a substrate on its own.[52] An industrial-grade version of the polymer with a thickness of ~200 μm was planarized with a thin polyamideimide layer ($n = 1.71$), and subsequently, a transparent electrode and a green phosphorescent OLED stack were deposited. Interestingly, industrial-grade PEN is not optically clear but shows a certain level of haze. The authors argue that this can be used to enhance the outcoupling by light scattering. Compared to a reference OLED on a conventional glass substrate, the efficiency was increased by 1.3-fold to ~30% EQE. It will be interesting to see how this concept translates to white OLEDs.

A very recent publication by Kim et al. elegantly demonstrated the limits of the above approaches, even though the study was performed only for monochrome OLEDs.[53] First, in order to reduce losses to metal surface plasmons to the greatest possible extent, the authors developed a metal-free device having transparent conductive oxides as both electrodes (Fig. 3). On the side of the substrate opposite to the device, a conventional MLA was applied. Unfortunately, polymer films comprising microlenses cannot be readily laminated on top of an OLED stack due to the risk of introducing local mechanical defects. To solve this problem, the authors deposited a very thick layer of organic material [in this example, N, N'-Di(napthalen − 1 − yl) − N, N'-diphenyl-benzidine (NBP)] through a metal shadow mask with micrometer-sized holes, such that a hexagonal array of cone-like structures was formed (~60 μm diameter and 80 μm height). This led to a 2.6-fold increase in EQE and luminous efficacy compared to a reference without microstructures. Despite the high values reached, it remains to be seen whether this approach is practical; it consumes way beyond 100 times

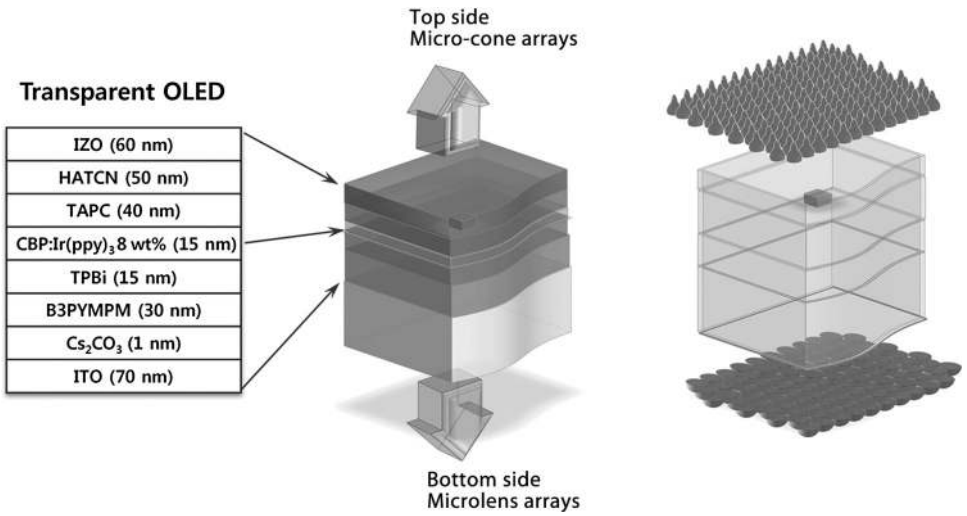

Fig. 3 Structure of a transparent OLED with attached micro-cones proposed by Kim et al. that is designed to have minimum plasmonic losses and to achieve efficient extraction of light from waveguided modes. These OLEDs can be 2.6-fold more efficient than the respective reference device. However, the concept has only been used for monochrome OLEDs so far. (Reproduced with permission, courtesy of John Wiley and Sons.)[53]

the organic material needed for the actual device (~100 nm compared to the 80 μm organic cone heights).

5 Wavelength-Independent, Scattering-Based Outcoupling Approaches

Given the practical issues associated with the high refractive index approach described above, there is strong motivation to develop alternatives for efficient extraction of organic and/or surface plasmon modes, in particular because the biggest fraction of power generated in the emission layer is confined to these modes [cf. Fig. 1(d)]. However, so far this remains challenging and it is important to note that the overall outcoupling efficiency of any specific device must be treated holistically, especially because many of the approaches that have been suggested cannot be combined in an additive fashion. Instead, the OLED under study has to be treated as a complete optical system for which the most promising routes of light extraction need to be specifically identified.

One general approach to enhance the light outcoupling from organic waveguide modes is the introduction of controlled lateral structures with the aim to redirect the light to the outcoupling cone. Sun and Forrest fabricated a low refractive index grid ($n_{\mathrm{grid}} = 1.45$) with dimensions on the micrometer scale between ITO and the organic layers, which allowed picking up and

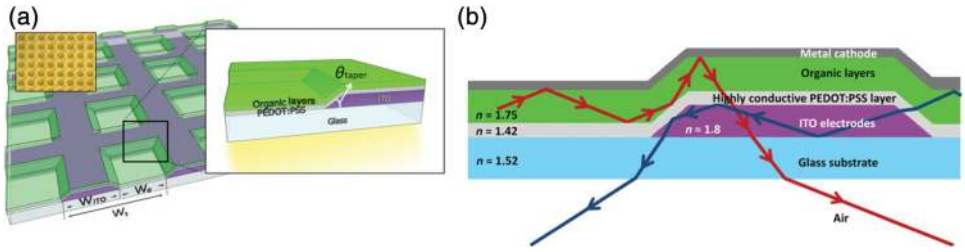

Fig. 4 Schematic illustration of an ITO grid-based approach to extract light from modes waveguided in the organic layers of OLEDs: (a) overview and (b) cross-sectional view indicating selected light paths. An ITO grid with tapered edges is coated with the low-index conductive polymer PEDOT:PSS to facilitate scattering of light guided in the organic film. Mesh size, line width, and taper angle can be tuned to optimize the efficiency of this approach. (Reproduced with permission, courtesy of John Wiley and Sons.)[54]

redirecting light traveling in the plane of the device.[45] When combined with the microlens films discussed in the previous section, a wavelength-independent improvement in outcoupling by a factor of 1.68 can be achieved. Koh et al. have suggested a similar concept based on a tapered ITO grid fabricated by conventional photolithography techniques.[54] To introduce a contrast in refractive index, the ITO structure is capped with the low refractive index conductive polymer poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) (Fig. 4). The outcoupling enhancement results from a complex interplay between guiding light in the plane and extracting the guided light via out-of-plane reflection by the special geometric shape of the ITO lines. For instance, for the former concept of using low refractive index grids, the largest improvement is observed when this approach is combined with an additional MLA; at best, the device efficiency can be enhanced by 1.68 times compared to the respective reference device.

An important consideration in the context of extracting waveguided and plasmonic modes is the decay length of these modes, i.e., the distance over which they propagate before their energy is lost to absorption. For a typical device structure, the decay length of plasmonic and waveguided modes is 2 and 20 μm, respectively.[40] For a scattering structure to be effective for a particular mode, photons trapped within this mode must be likely to be scattered before they are lost, i.e., the modal scattering length (=scattering length × overlap of mode with scattering structure) has to be smaller than or comparable to the decay length of the modes that are to be extracted. This requires large scattering coefficients and close proximity of the scattering structure to the active layers of the OLEDs, in particular for plasmonic modes. Figure 5 shows mode profiles of the lowest-order waveguided and plasmonic mode supported by a standard bottom-emitting OLED stack and how these modes can interact with a scattering structure located underneath the anode of the device.[40] Chang et al. have demonstrated that polymer films filled with TiO_2 nanoparticles can serve as efficient scattering layers in this configuration and that the approach is compatible with both ITO (Refs. 40 and 55) and PEDOT/PSS anodes.[56] Recently, Li et al. combined the nanoparticle scattering approach with flexible nanocomposite electrodes comprising single-walled carbon nanotubes (CNTs) and silver nanowires, again leading to a considerable increase in outcoupling efficiency.[57]

The methods discussed above make use of structures that scatter light incoherently. By reducing the size of the features further and introducing periodicity, one can make beneficial use of coherent effects, in particular of Bragg scattering of the trapped light by grating-type structures.[58–60] However, in general, these photonic structures are wavelength selective, which is

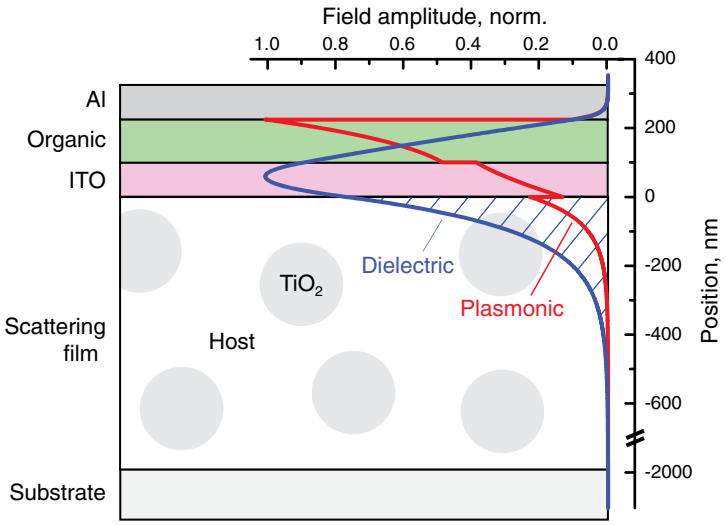

Fig. 5 Schematic illustration of how nanoparticle-based scattering layers can enhance light extraction in OLEDs. A scattering film is inserted between the substrate and the OLED stack. The blue and the red lines represent the field distribution of the lowest-order s-polarized dielectric mode and the plasmonic mode supported by the structure (for $\lambda = 520$ nm). The overlap between the evanescent tail of these modes and the scattering layer facilitates extraction of otherwise trapped light. (Reproduced with permission, courtesy of AIP Publishing.)[40]

why they are difficult to apply in broadband white OLEDs. In order to circumvent the wavelength selective character, attention has been drawn to quasi-periodic structures that offer efficient scattering across the full range of the visible spectrum.

One way of producing such quasi-periodic structures has been demonstrated by Koo et al., who formed subwavelength corrugated modulations in the substrate by cooling a composite system of thermally expanded PDMS and aluminum. Due to differences in the thermal expansion coefficients, buckling structures are created instantaneously, allowing scatter guided modes to the outcoupling cone via Bragg scattering.[61] So far, this concept has only been tested for single-emitter OLEDs; however, the used material covered a large fraction of the visible spectrum (substantial emission from 475 to 675 nm). The enhancements observed by the authors range from roughly twofold in the blue region to three- to fourfold in the red part of the spectrum (depending on the actual condition of the buckling structure underlying the device). Even though the enhancement factor thus shows some wavelength dispersion, quasi-periodic buckling structures appear to be a valid concept for white OLEDs. The remaining wavelength dependence could be counteracted by iteratively redesigning the exciton distribution between the various emitters.

Ou et al. applied the concept of random structures to both the back and the front side of OLEDs via consecutive nanoimprint steps.[62] First, a poly fluoro poly ether master stamp is used to create concave deterministic aperiodic nanostructures (DANs) in a PEDOT:PSS

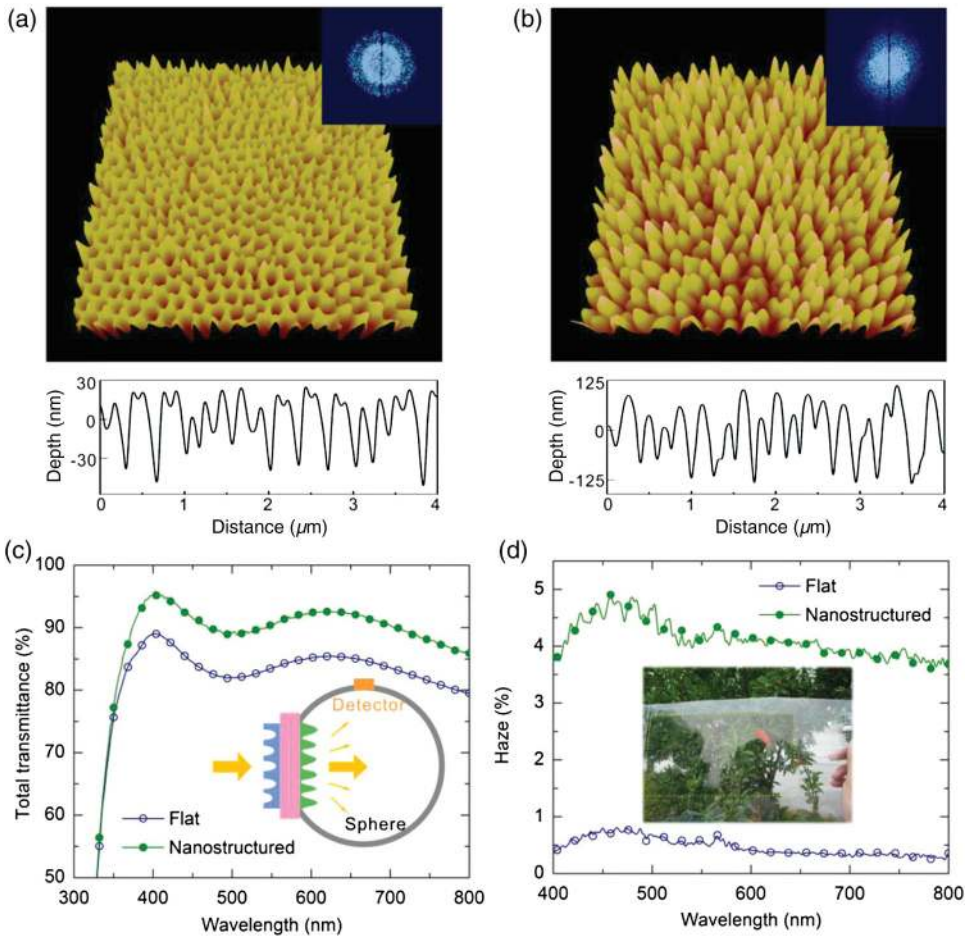

Fig. 6 Characteristics of deterministic aperiodic nanostructures (DANs) developed by Ou et al. and fabricated by nanoimprint processes.[62] (a) and (b) AFM images of the back and front side structures, respectively, and corresponding depth profiles, (c) transmission through back and front DANs held by a center substrate and transmission of flat reference as a function of wavelength, and (d) the respective haze values of nanostructured and flat substrates. (Reproduced with permission, courtesy of John Wiley and Sons.)[62]

layer that was fabricated onto an ITO/glass substrate. On top of the modulated PEDOT:PSS, the complete OLED is deposited in a conformal way, creating an internal DAN. After device fabrication, the substrate is flipped around and another master stamp is used to generate an external, convex DAN into an additional layer sitting remotely on the glass side opposite to the device. The resulting topographies of internal and external DANs are shown in Fig. 6. Overall, the combination of internal and external DANs allows obtaining an external quantum efficiency of ~54% (123 lm/W; 2.1-fold increase over the optimized flat reference device). This is one of the highest values reported for white OLEDs so far, thus clearly demonstrating the effectiveness of the DANs used.

Another elegant way to enhance the outcoupling of waveguided light in a wavelength-independent manner is to insert scattering structures directly into the OLED layer stack. This is very challenging because the introduction of scattering particles and/or layers is likely to lead to structural defects in the device and, thus, can dramatically increase leakage current. Löser et al. (Novaled AG) reported on a device structure that comprises an electron transporting material (called NET-61) that forms nanocrystallites when embedded into an OLED stack and thus, effectively constitutes an internal scattering layer.[63] When introduced in a bottom-emitting triple stacked white OLED (as the electron transporting layer of the cathode-facing unit) and combined with an additional external microlens film, this yields a 1.85 times higher external quantum efficiency compared to an optimized reference. Device stability was not affected by the introduction of NET-61.

6 Microcavity-Based Approaches

Most efforts to develop novel outcoupling schemes for OLEDs concentrate on bottom-emitting devices. This comes as no surprise because (1) the cavity of the device is much weaker, making it optically more robust and (2) modification of top-emitting OLEDs is far more challenging as it has to be done after the deposition of the soft organic layers forming the OLED stack. Top-emitting OLEDs often comprise metal layers for both anode and cathode (typically one reflective, one semitransparent), which leads to a cavity with a rather strong wavelength selectivity. Semitransparent metal layers are often the only option for the top contact as deposition of other transparent electrode materials (e.g., transparent conductive oxides, typically deposited by sputtering) tends to harm the underlying, soft organic layers. A common problem for white top-emitting OLEDs that results from the strong microcavity is that certain wavelength regions within the visible spectrum are suppressed while others are enhanced. Thus, while high external quantum efficiency values can be obtained, one can hardly achieve a broad, balanced white spectrum. In addition, white top-emitting OLEDs comprising strong cavities show a very pronounced angular dependence of the emitted light. While devices that change color with the viewing angle may be of interest for decorative lights (e.g., Christmas tree lights), they do not qualify for most solid-state lighting applications.

For monochrome devices where the desired emission is only within a narrow sub-band of the visible spectrum, strong cavities can be used to enhance the external quantum efficiency.[64] Here, outcoupling enhancements from 1.2- to 1.5-fold have been observed, with increasing enhancement factors observed for increasing device resonance wavelength. By adjusting the optical cavities very carefully and with the aid of optical simulation, it is even possible to fabricate white OLEDs that are composed of three stacked microcavities with specific resonances in the blue, green, and red.[65] While the overall efficiency of such a multicavity design can be very high, it still lacks angular stability of the emitted light.

The simplest possible route to reduce the wavelength selectivity of top-emitting OLED cavities is to add an organic (dielectric) layer on top of the semitransparent electrode. Such a passive layer with exclusively optical functionality is known as a dielectric capping layer. Capping layers have been successfully used to design highly efficient monochrome[66–68] and even white OLEDs.[69] With a capping layer, the optical properties of the complete multilayer device can be significantly altered. For outcoupling reasons, it can be tailored to function as an antireflection coating, weakening the cavity strength, slightly improving the light outcoupling efficiency, and, therefore, allowing broadband emission with a reduced angular shift to be

emitted from white top-emitting OLEDs. Similarly, a capping layer can be designed to increase the reflectance of a device stack, consequently enhancing cavity resonances that may be desired in other applications.

Thomschke et al. solved the problem of the high degree of wavelength selection in an elegant, yet unconventional way.[70] They fabricated two-unit stacked OLEDs, hosting green and blue/red emission layers in the respective stacks, and optimized these for maximum external quantum efficiency disregarding the poor color quality of the resulting device. After applying an organic capping layer, they laminated a high refractive index MLA film onto the device. Similar to the evaporated micro-cones reported by Kim et al.,[53] this yielded a device that comprises an outcoupling layer on top of the top-most organic layer and that does not show any substantial refractive index contrast throughout the stack. The MLA simultaneously functions as mode mixing

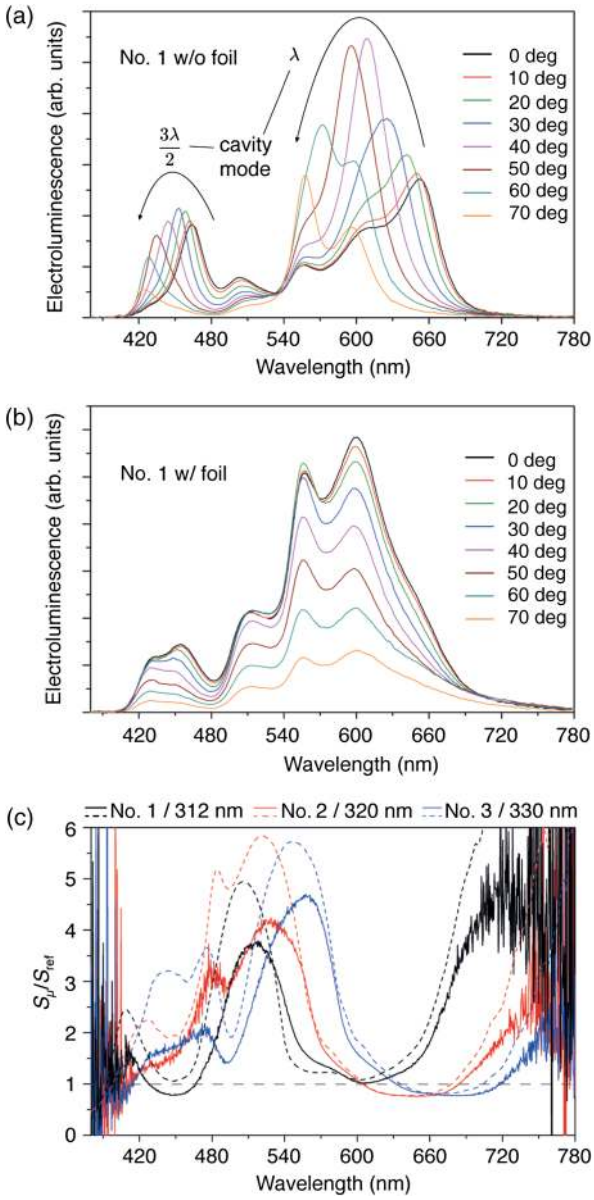

Fig. 7 Emitted electroluminescence spectra as a function of angle of observation for devices (a) without and (b) with attached microlens array (MLA). (c) The spectral outcoupling factor of the MLA with respect to the device without MLA. In the green part of the visible spectrum, the MLA strongly contributes to the outcoupling of trapped waveguided modes, which are suppressed in the reference device [cf. (a)]. (Reproduced with permission, courtesy of American Chemical Society.)[70]

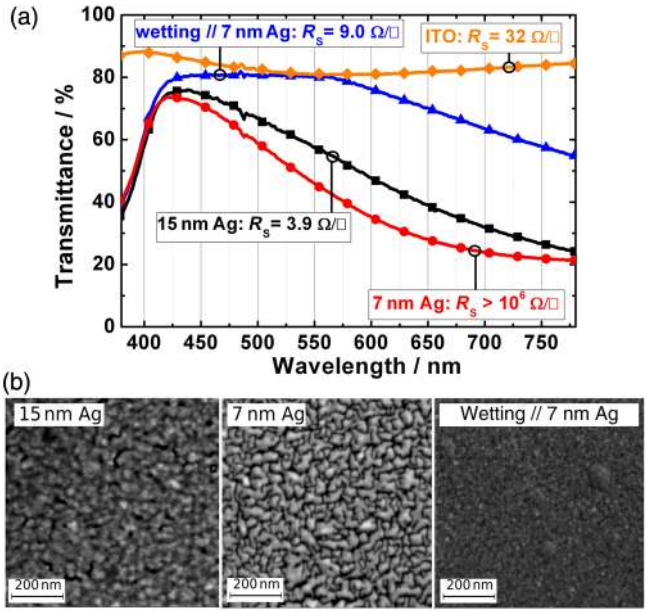

Fig. 8 Comparison of different electrode systems. (a) Optical transmittance of a reference ITO electrode, 15 and 7 nm silver layers, and 7 nm silver layer grown on an additional wetting layer. Clearly, the composite electrode (wetting +7 nm silver) outperforms the thicker silver-only electrodes and shows a performance comparable to ITO having high transmission and low sheet resistance. (b) SEM images of the three different silver electrodes. Plain layers show significant island growth, while the composite approach results in uniform, smooth, and closed layers. (Reproduced with permission, courtesy of John Wiley and Sons.)[71]

and mode extracting systems, thus facilitating white top-emitting OLEDs with angle-independent emission color, well-balanced emission spectra, and enhanced external quantum efficiency. Figure 7 shows the respective spectra as a function of observation angle for devices without and with applied MLA, illustrating the mode mixing and outcoupling enhancing properties of the laminated MLA. This concept should be easily scalable; the process may even be compatible with roll-to-roll processing techniques.

Recent work on white top-emitting OLEDs has employed alternative, improved top electrodes that combine high optical transmission, high lateral conductivity at least on par with ITO, and good process compatibility. Schwab et al. focused on metal electrodes. Conventionally, at least 15 nm of silver are needed on top of the organic layer stack to form a closed metal film with sufficient conductivity. At this thickness, silver films have significant reflectance and absorption (transmittance only 30 to 75%, depending on wavelength).[60] By introducing a multilayer electrode comprising a wetting layer to support silver growth and an ultrathin silver layer, very attractive conductivity and transmittance are achieved with much thinner silver layers, down to 5 nm in thickness.[71,72] Transmission data of these ultrathin, composite electrodes are compared to ITO and thicker films of pristine silver in Fig. 8. With these ultrathin, highly transparent, and highly conductive composite electrodes, both high external quantum efficiencies (comparable to bottom-emitting reference devices) and emission spectra that are stable over a wide range of observation angles can be achieved.[71,72] With their high transparency, these ultrathin electrodes pave the way to the ultimate design freedom for OLEDs. The low temperature processing conditions allow positioning them at any location within a device stack. For instance, such electrodes can be used as highly transparent, yet conductive interconnects that enable device stacking and, consequently, color tuning without sacrificing device efficiency due to the added electrodes.[73]

Alternatively, as reported by Freitag et al., CNT meshes can be used as highly transparent, conductive top electrodes for white OLEDs.[74] Here, freestanding CNT meshes with a sheet resistance of 1500 Ω/□ were laminated onto the organic layers. Due to the low CNT conductivity, the device power efficiency falls short by roughly one order of magnitude compared to a reference device comprising a 15 nm silver electrode. However, a key advantage of this approach is its optical design. The CNT meshes greatly reduce the microcavity simply because most of the

device area is actually not covered by the CNTs.[74] Consequently, the emitted spectra remain virtually unchanged across all angles of observation. In the future, the combination of CNT meshes with metal support grids may alleviate the issues associated with their low conductivity.

7 Emitter Orientation

The orientation of the emissive molecules within an OLED stack strongly affects the outcoupling efficiency. In this final section of our review, we will discuss some recent work aimed at exploiting orientation effects to further increase the efficiency of light extraction.

In 2000, Kim et al. showed that for polymer-based OLEDs, the emissive dipoles tend to be aligned horizontally, i.e., within the plane of the device, and that this is expected to lead to a higher outcoupling efficiency than possible for a random isotropic orientation of dipoles.[33] Due to the donut-shaped radiation pattern generated by an emitting dipole, a dipole that is aligned vertically with respect to the plane of the device generates very little emission at low k_x/k values, i.e., only a small fraction of the generated light falls into the outcoupling cone. Instead, most of the power from a vertical dipole is coupled to substrate, waveguided, and plasmonic modes. A dipole with horizontal orientation instead emits a considerably larger fraction of its power into the air surrounding the device. The long and rigid shape of polymers, combined with the fact that the transition dipole moments of polymers tend to be aligned along the polymer chain,[75] means that a preferential horizontal dipole orientation is frequently obtained for these materials.

By contrast, it has long been argued that the advantageous light outcoupling enhancement of oriented emissive dipoles cannot be harnessed for small molecule-based OLEDs because they were believed to be isotropically oriented. However, research on linking molecular orientation to enhanced outcoupling efficiency recently gained new momentum through work by Yokoyama et al., who showed that linear-shaped organic molecules with suitable substituents can spontaneously align within a neat thin film deposited by thermal evaporation in high vacuum.[76] Yokoyama also hypothesized that this alignment could yield substantial efficiency improvements in state-of-the-art small molecule OLEDs. Flämmich et al. then reported on a more accurate method to measure the orientation of emissive dipoles in an electrically driven OLED.[77] Shortly thereafter, Frischeisen et al. demonstrated that in fluorescent, small molecule OLEDs, horizontal emitter orientation can indeed increase outcoupling efficiency. In this first example, 88% of the dipoles were estimated to have horizontal orientation (66% would correspond to isotropic orientation), and a comparison between an aligned and a nonaligned dye indicated that the horizontal orientation led to a 1.45-fold efficiency increase.[78]

While this first work was based on elongated fluorescent emitter molecules, it is less obvious that phosphorescent emitter complexes, which tend to have a more bulky molecular structure, can also show preferred molecular orientation. However, applying angle-resolved and time-resolved measurements, respectively, Flämmich et al.[79] and Schmidt et al.[80] have found evidence that the emission from OLEDs based on the red phosphorescent emitter $\mathrm{Ir(MDQ)_2(acac)}$ is best modeled by emissive dipoles with preferential horizontal orientation. Figure 9(a) shows measured and calculated angle-resolved emission spectra of these $\mathrm{Ir(MDQ)_2(acac)}$ OLEDs and illustrates how comparing experimental data to different simulated datasets facilitates an accurate measurement of the average emitter orientation. For the phosphorescent $\mathrm{Ir(MDQ)_2(acac)}$ OLEDs, a smaller fraction of the molecules is oriented horizontally (76%) than in the earlier fluorescent devices, and consequently, the improvement in outcoupling compared to a situation with isotropic emitter orientation is less pronounced. Nevertheless, theoretical calculations indicate that with further improvements in orientation, outcoupling efficiency can be enhanced 1.5-fold compared to the isotropic case [Fig. 9(b)]. In addition, emitter orientation could be combined with many of the other outcoupling schemes described above, even though the effects may not be fully additive.

Subsequent work by Liehm et al. compared the emission of the two structurally similar green emitters $\mathrm{Ir(ppy)_3}$ and $\mathrm{Ir(ppy)_2(acac)}$ and established that the higher EQE observed in $\mathrm{Ir(ppy)_2(acac)}$-based OLEDs is to a large degree due to preferential horizontal orientation[81] although aggregation effects are also important.[82] Since then, there have been several reports demonstrating that the efficiency of phosphorescent OLEDs can be enhanced by exploiting orientation effects.[83,84] However, it remains challenging to obtain the same level of molecular

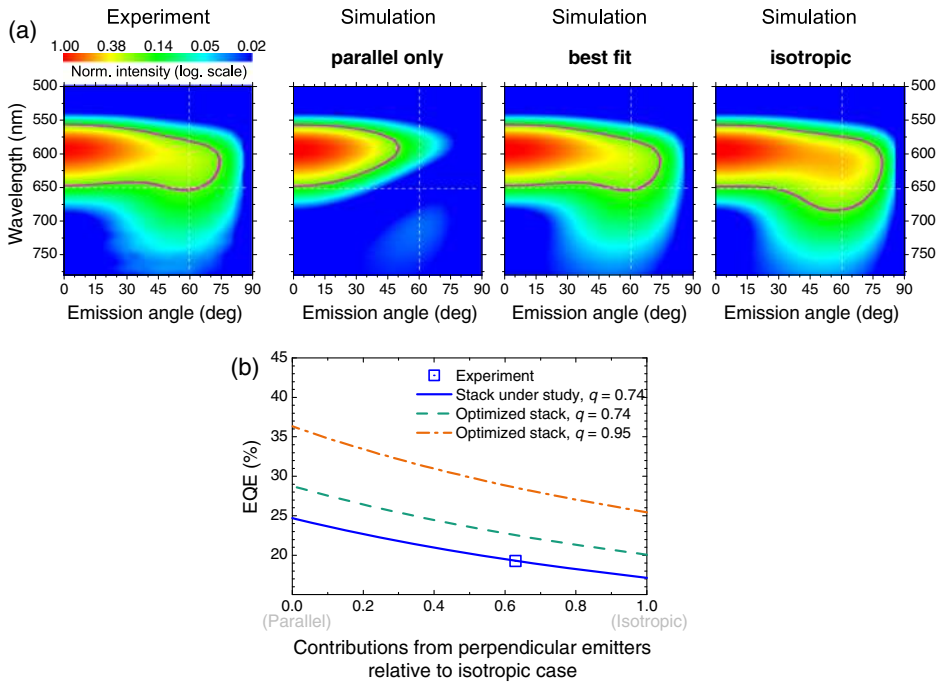

Fig. 9 (a) False color plots of the experimental and theoretical angle-resolved emission spectra of an $Ir(MDQ)_2(acac)$-based OLED designed to show strong changes in emission characteristics with emitter orientation (electron transport layer thickness, 160 nm). The magenta line indicates a normalized intensity of 0.2 to enable better comparison between experiment and the three different simulations, each of which is performed for a different average emitter orientation. Neither the situation of parallel only dipoles nor the isotropic case describes the data whereas the best fit (76% orientation) is in good agreement with the experiment. (b) Optical simulation of expected external quantum efficiency as a function of the degree of emitter orientation for a red phosphorescent OLED. Different stack designs and different photoluminescence quantum yields q are considered. (Reproduced with permission, courtesy of Elsevier.)[79]

orientation for phosphorescent emitter molecules that has been achieved with some of the best fluorescent emitters. In fact, the considerable efficiency increases demonstrated in Refs. 83 and 84 are due to a combination of orientation effects and an increased photoluminescence quantum yield of the new, more highly oriented emitters.

Research aimed at understanding why certain phosphorescent emitters show preferred horizontal orientation despite having a bulky rather than an elongated structure has shown that van-der-Waals type interactions between emitter molecules and with matrix molecules are important.[84,85] Kim et al. have performed quantum chemical calculations to estimate the orientation of the transition dipole moment with respect to the relevant axes of the molecule and also to determine the supramolecular arrangement of four different emitters within a host environment (Fig. 10).[84] Graf et al. have carried out a comprehensive study that compares the orientation of seven iridium-based phosphorescent emitters and found that with the exception of $Ir(ppy)_3$, all studied emitters have a pronounced tendency for horizontal dipole orientation.[85] Using quantum chemical calculations again, evidence was obtained that—at least for the emitters in this study—orientation is linked to the dipole–dipole interaction potential between molecules and hence possibly to the tendency of the emitters to form aggregates.

Emitter orientation has also been investigated for the emerging class of TADF emitters, and first results indicate that for large, planar TADF molecules, very high levels of molecular orientation may be feasible, with >92% of the molecules being aligned horizontally in one particular case.[86]

While the above work impressively demonstrates that emitter orientation can enhance outcoupling efficiency, there has, to the best of our knowledge, so far been no work on white OLEDs that exploit emitter orientation in a directed manner. However, highly efficient phosphorescent

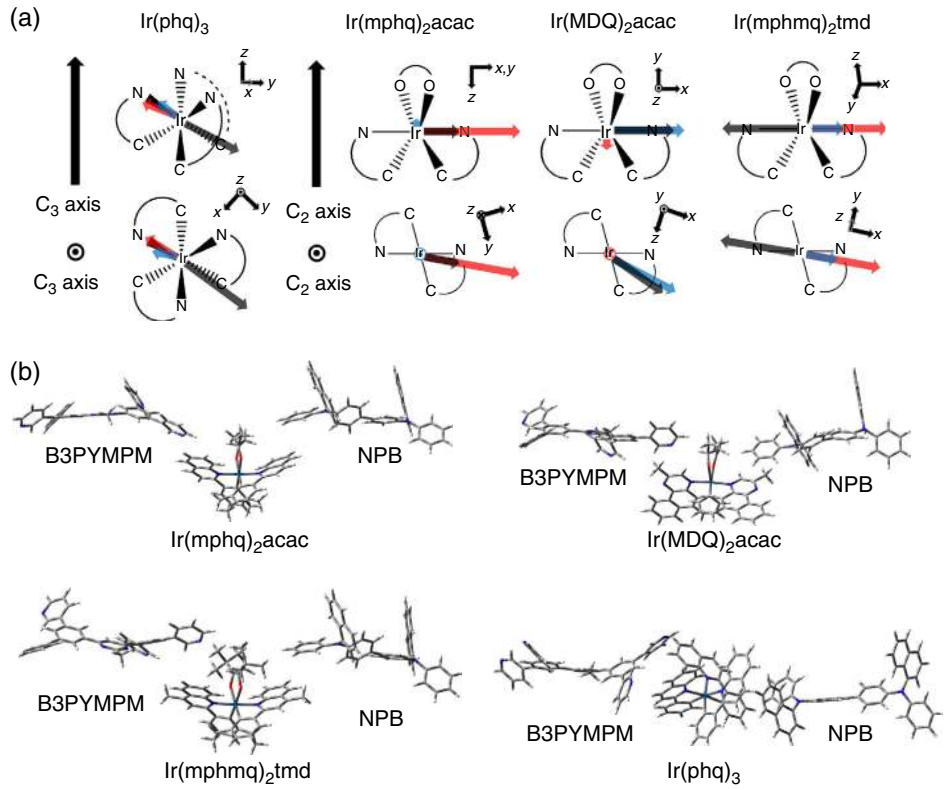

Fig. 10 (a) Orientation of the triplet transition dipole moments of four different red phosphorescent emitters as obtained by quantum chemical calculations. The dipole moments of the three different triplet sublevels are indicated by black, red, and blue arrows, respectively. (b) Optimized binding geometries of the same emitters within a bimolecular host comprising bis{4,6-[3,5-di-(3-pyridyl)]-2-methylpyrimidine} (B3PYMPM) and NPB. (Reproduced with permission, courtesy of Nature Publishing Group.)[84]

emitter molecules spanning the entire visible spectrum are now available,[85,87,88] and their horizontal orientation may, in fact, have already been harnessed, even though inadvertently.

Applications of OLEDs in general illumination require devices to operate at high brightness levels where roll-off effects are often important, i.e., the efficiency of an OLED operated at brightness levels relevant for general illumination (e.g., 5000 cd/m^2) may be considerably less than the efficiency at low brightness (e.g., at 100 cd/m^2).[89] Murawski et al. have recently found that molecular orientation feeds back into these roll-off effects due to an intricate interplay of different effects.[90] Horizontally orientated dipoles couple less strongly to surface plasmon and waveguided modes. The reduced coupling leads to a smaller Purcell factor and, thus, to a longer excited state lifetime than for isotropically orientated emitters. This, in turn, leaves more time for bimolecular annihilation processes, in particular triplet-triplet annihilation, and thus causes increased roll-off. Consequently, for very high brightness applications, there may be a trade-off between improved outcoupling efficiency and the increase in efficiency roll-off due to triplet-triplet annihilation.

8 Conclusion

In conclusion, a range of different loss channels and effects play significant roles in determining the outcoupling efficiency of white OLEDs. So far, only some of these losses can be addressed in an efficient and practical manner; e.g., a substantial degree of substrate-guided light can be recovered by external outcoupling structures. For other loss channels, there have been impressive proof-of-concept demonstrations, but further work will be required to make the approaches fully compatible with large-scale production in terms of cost, yield, device stability, etc. The details of some loss processes and how they are influenced by the outcoupling methods currently available

are not yet fully understood. In particular, future research should be performed to obtain a more complete understanding of coupling to surface plasmon modes and to identify generally applicable approaches for its elimination. Another priority of future research should be to study in more detail the effects induced by molecular orientation and to identify ways of controlling orientation in efficient emitter materials.

At present, a multitude of different outcoupling approaches exist, and it is important to realize that many of these cannot be combined in an additive manner. Therefore, a holistic approach is required, taking into account the different options and various degrees of freedom in device design, with the goal of obtaining the best possible light extraction efficiency without affecting device stability, color quality, or compatibility with flexible substrates. This can only be achieved if the success of new outcoupling concepts is measured by comparing them to optically optimized conventional devices based on the same emitters and to devices that comprise one or several of the more established outcoupling schemes.

Acknowledgments

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