Improve OLED Light Outcoupling Efficiency by Eliminating Waveguide Mode using Ultrathin Metal Electrode

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Abstract
Extracting light from the waveguide mode in organic light-emitting diodes (OLED) is most challenging as such optical mode is intrinsic in traditional OLED due to materials and layered structure used in the device. This work intends to address the root cause of the light trapping problem. We systematically analyzed OLEDs made with an ultrathin Ag anode and found that waveguide modes can be completely eliminated by designing a structure to below the cutoff thickness of waveguide modes. We also experimentally verified the waveguide mode elimination in organic waveguides. In principle such a scheme can be extended to tandem cells, and in making transparent OLED. A further benefit is the ductility of the ultrathin metal allows foldable OLED to be realized.

Author Keywords
Organic light-emitting diodes (OLEDs); metal-based transparent conductor; anti-reflection; outcoupling; waveguide mode.

1. Objective and Background

Organic light emitting diodes (OLEDs) have been widely used in flexible and transparent display products (1). Despite the success in material progress and achieving 100% internal quantum efficiency (IQE), external quantum efficiency (EQE) of OLED is still limited due to the poor outcoupling efficiency (2). There has been strong demand to out-couple the trapped light in OLED for higher external quantum efficiency, and hence intensive research. Multiple solutions have been proposed, including the introduction of ITO-free OLEDs that improved device efficiency.

There are three major factors that limit the EQE of OLEDs: (1) Generated photons in a device are lost to the contact metal due to excitation of plasmons that do not radiate—the effect can be reduced by resorting to thick electron transporting layer (ETL) (4-6) (2) Emitted light total-internally reflected from a glass substrate—this can be reduced by using microlens array (5) or light scattering structures in the substrate (6). (3) Light trapped in a device in the form of guided mode due to the device layers serving as waveguide (2, 3)—this is the hardest to extract, because the guided mode formed within the active region including the transparent anode is inherent and propagates along these layers. Especially when sufficiently thick layer of transparent conductive oxides (TCO) as an anode is needed to ensure low film resistivity, it aggravates the waveguide effect.

There have been efforts to extract the waveguide mode, such as by patterning or using grid structure, but they not only increase the fabrication complexity and cost, but also easy to protrude into organic layers or results in non-planar surfaces, negatively affecting the surface smoothness important for OLED and its operation lifetime (6, 7). In this work, we exploit a simple yet effective solution to eliminate the waveguide mode and increase EQE without adding fabrication cost or affecting other properties of the OLEDs.

2. Results and Discussion

Mechanism of waveguide elimination: Our approach is to use an ultra-thin Ag layer, e.g. < 6 nm, to replace the traditional ITO electrode. Such a simple replacement has an immediate effect on the optics in OLED: waveguide modes are no longer supported in the OLED layers. We will use Fig. 1 to explain. The schematic of OLED showed the waveguide mode TE_0 and TM_1, which are the two lowest order modes typically guided in traditional ITO based devices. This is because ITO has the highest refractive index of all layers. In contrast, low-loss metal such as Ag has real part of the refractive index close to zero, therefore can drastically reduce the effective index (N_{eff}) of the waveguide layers. When N_{eff} is reduced to the that of the substrate, waveguide cannot be supported. N_{eff} was calculated for TE_0 (Fig. 1b) and TM_1 mode (Fig. 1c) for OLEDs made with ITO anode (black) and thin-Ag anode with varied thickness (red, solid or dotted lines), as well as device without an anode (blue). Clearly the introduction of thin Ag film imposes waveguide cutoff, but the cutoff thickness is close to 200nm for TE_0, which is sufficient thick for most of the OLED structures. This means that no other change needs to be made to a high-performance OLED. Increasing the thickness of the Ag anode can help to increase the cutoff thickness (arrow direction in each figure), but with trade off lower transparency and potential micro-cavity effect due to stronger reflection from the Ag film. TM_1 mode has similar property, but with reduced cutoff thickness of the OLED layers with increasing Ag thickness; while ITO device has cutoff well below 100nm, which is impractical to realize. Considering both TE and TM modes, ideally thin Ag thickness should be < 10 nm for mode elimination.

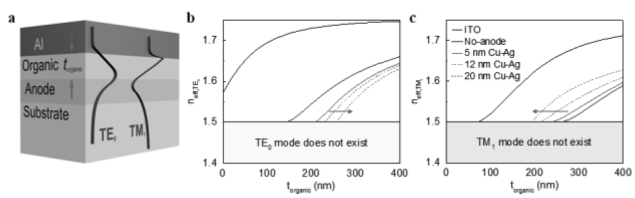

Figure 1. (a) Schematics of the calculated waveguide structure. Electric field of TE_0 mode and magnetic field of TM_1 mode were also depicted. Calculated effective indices of (b) TE_0 ($n_{eff,TE0}$) and (c) TM_1 ($n_{eff,TM1}$) modes as functions of organic layer thickness $t_{organic}$.

Fabrication of ultra-thin and smooth Ag film: The apparent electrical resistivity of a very thin Ag film becomes strongly dependent on the film's thickness, and a rapid change in ultrathin metal film's electrical and optical properties as the thickness shrinks to below certain critical thickness (Fig. 2a), understanding the resistivity change at ultrathin regime may provide insight and guidance in engineering applications. We recently showed that its maximum theoretical figure-of-merit (FOM defined as the ratio of optical transmittance over sheet resistance) is determined at the critical thickness, and serves as an

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important engineering metric (8).

In order to obtain continuous, smooth ultrathin Ag films required for OLED, one needs to overcome the intrinsic Ag de-wetting problem. With appropriate seed-layer, we found that the Ag layer can be controlled to have 2D-like growth with low electrical resistance and optical loss. The optimum thickness of Cu seed-layer was ~5 Å, which guarantees ultrathin and smooth Ag film with low electrical resistance and high transmittance.

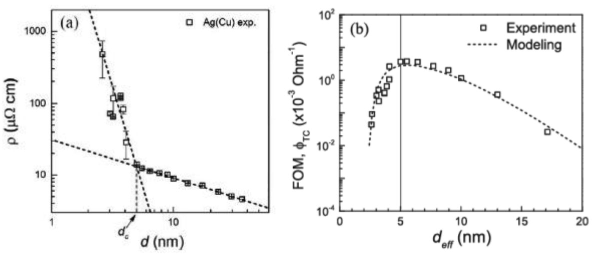

Figure 2. (a) Resistivity ρ of Cu-seeded Ag (Cu) film as a function of film thickness d in log-log scale. (b) Haacke's figure-of-merit ϕ_{TC} (= T^{10}/R_s) of Cu-Ag film as a function of d_{eff} where T is transmission at 550 nm wavelength and R_s is sheet resistance.

Modal power distribution in OLEDs: Optical property of OLEDs with the thin Cu-Ag anode is investigated and compared to that with ITO anode. Here a typical organic structure was used for the calculation, where the stack consisting of 5 nm molybdenum trioxide (MoO_3) / 40 nm di-(4-(N,N -di-p-tolyl-amino)-phenyl) cyclohexane (TAPC) / 20 nm EML / 75 nm 1,3,5-Tris(1-phenyl-1Hbenzimidazol-2-yl)benzene (TPBi), where CBP was used for the EML in the calculation. Figures 3a and 3b show spectral power distribution of each mode in the ITO and thin Cu-Ag based OLEDs, respectively, which was calculated by Dyadic Green's function. The ITO device showed relatively similar amount of the power distributed in each mode, with the highest fraction at the substrate mode. In contrast, the waveguide mode (W/G) in Cu-Ag device has completely disappeared within the entire emission wavelength range, and this power is distributed to the other modes. Among them, the substrate mode showed the largest increase, which now can be extracted, e.g. by using microlens or scattering surface. (5,6).

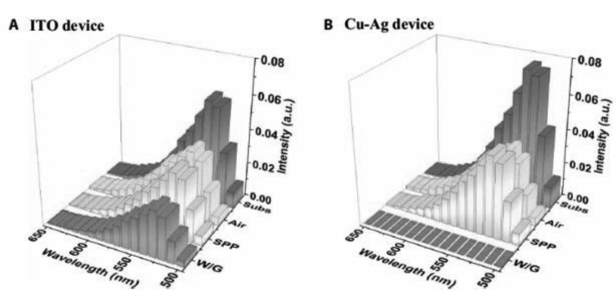

Figure 3. (a) Schematics of the calculated waveguide structure. Electric field of TE_0 mode and magnetic field of TM_1 mode were also depicted. Calculated effective indices of (b) TE_0 ($n_{eff,TE0}$) and (c)

Results of fabricated OLEDs: A green OLED based on a conventional structure was fabricated with either ITO or Cu-Ag anode to observe the effect of the waveguide mode removal on the device outcoupling efficiency. Fabricated OLEDs consist of glass substrate / 150 nm ITO or 5 nm Cu-Ag / 5 nm MoO_3 / 40 nm TAPC / 20 nm 10% $Ir(ppy)_2acac$ doped in CBP / 75 nm TPBi / 1.5 nm LiQ / 150 nm Al, where OLEDs with ITO anode serves as a reference. Figure 4a shows measured EQE versus current density characteristic of each device. EQEs of the ITO and Cu-Ag devices were 20.2% and 19.9%, respectively, which are similar fraction to the calculated air mode portion shown in Figures 3.

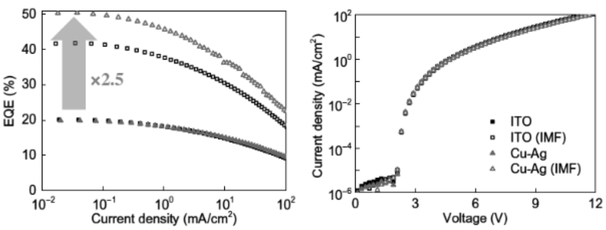

Figure 4. (a) EQE – current density and (b) current density – voltage characteristics of Cu-Ag and ITO OLEDs with and without IMF. IMF was used to extract the light trapped in the substrate.

Experimentally the most straightforward method to extract the trapped light in the substrate is the use of index matching fluid (IMF). In practice, microlens array or other patterned structures can be used to achieve similar effect. The EQE of the Cu-Ag device significantly increased to 50.6% with the substrate mode extraction, whereas that of the ITO device remains maximum of 42.3% as shown in Figure 4a. The enhancement factor is among the highest ever reported. This efficiency improvement due to the substrate mode extraction showed the same trend with Figures 3. Figure 4b shows measured current density – voltage characteristic of the ITO and Cu-Ag devices. The two devices showed very similar electrical properties, overlapping in the curves. This indicates that electrical property of OLEDs was unaltered by replacing ITO with Cu-Ag. Therefore, the EQE enhancement from Cu-Ag is purely an optical effect and the result of more emitted photons being coupled out of the OLEDs.

Table 1 Comparison of the selected high-performance ITO-free OLEDs

Anode	R_{sh} (Ω/sq)	Control device EQE (%)†	Improved device EQE (%)‡	Enhancement Factor*	Reference
Cu-Ag	21.3	20.2	50.6	2.5	This work
PEDOT:PSS	145	26.4	64.5	2.4	(11)
PEDOT:PSS	145	23.1	44.1	1.9	(26)
PEDOT:PSS	150	14.1	17.2	1.2	(27)
Graphene	92.5	27.4	64.7	2.4	(24)

† Control EQE is the maximum EQE of the ITO reference sample on the flat glass substrate.
‡ Improved EQE is the maximum EQE of the suggested structure with the substrate mode outcoupling.
* Enhancement factor is the ratio between the control and improved EQEs.

We would like to point out that there is much interest in top-emitting OLEDs in industry that do not suffer from substrate loss. In such a structure, light extraction becomes more straightforward since the generated light can directly escape the device without the need of substrate mode extraction. Formation of waveguide mode is also suppressed in top-emitting OLEDs when thin Ag is used as transparent electrode. From waveguide point of view, glass clad is replaced with air in top-emitting OLEDs, which increases the refractive index difference between the waveguide core and clad thus requires thinner organic waveguide to achieve the waveguide mode cutoff.

Direct evidence of elimination of guided mode: The above results were consistent with an experiment we conducted earlier in polymer light emitting diodes, which were hypothesized due to the suppression of ITO waveguide mode (9). To provide a further direct experimental verification of the waveguide mode elimination, in this work we prepared organic waveguide samples grown on the ITO and Cu-Ag films. Strip-line organic layers with commercial ITO on a glass substrate and sputtered Cu-Ag on a fused silica substrate were prepared. By optical pumping the emissive layer and measure the light energy at the edge of the glass substrate, we clearly observed absence of waveguide mode in the Cu-Ag device.

3. Use of DMD with thin Ag for higher efficiency

Dielectric-metal-dielectric (DMD) based flexible transparent conductor shows one of the highest figure-of-merit as compared with other counterparts, indicating its great potential for flexible optoelectronic device application (10). The simplicity in process fabrication and compatibility with existing display technology makes it an excellent candidate for immediate use in industry. Especially the ultrasmooth surface of the DMD makes it ideally suited for the thin-film based OLED structures. In this regard, the use of Cu-doped or Cu-seeded Ag is important to guarantee a continuous nm thick Ag layer with smooth morphology.

We conducted optimization of DMD for optical transmittance and show that the optimum design for DMD used in OLED is different from that DMD in air. By this deliberate design of DMD, we demonstrate that the OLEDs using DMD electrode can outperform ITO-based counterparts. Moreover, we show that DMD based OLED on a flexible substrate can be bended to 2 mm radius of curvature and maintain normal operation. This work demonstrates that DMD is a good candidate to replace ITO for future flexible displays.

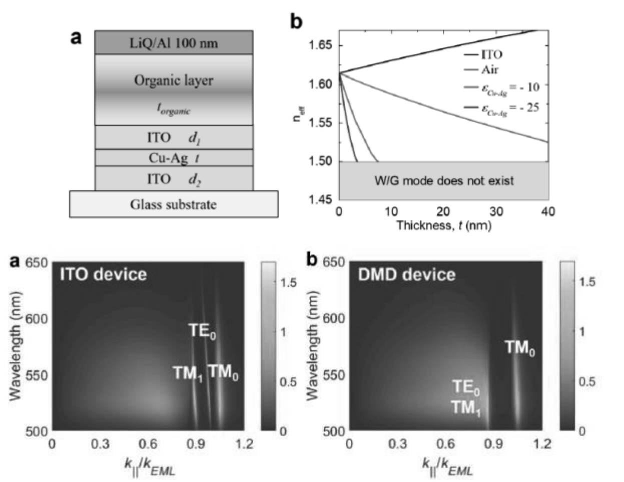

Figure 5. (upper panel) Modal analysis of an OLED with DMD. (a) Schematics of a DMD based OLED. (b) Effective index of TE_0 mode as a function of the Cu-Ag thickness with varied permittivity, with top and bottom ITO to be both 40 nm. ITO and Air in (b) indicate DMD with ITO and air permittivity instead of Cu-Ag. (lower panel) Spectral power distribution of an ITO or DMD OLED.

Typical high-performance OLEDs have organic thickness ($t_{organic}$) below 150 nm. As shown in Figure 5 the cutoff condition for the guided mode with DMD can still be satisfied for practical OLED applications.

Spectral power distribution in the ITO or DMD based OLED with dipole source is also investigated in Figures 5a and 5b (lower panel), respectively. The OLED structure used in the calculation is 5 nm MoO3 / 40 nm TAPC / 20 nm EML / 50 nm TPBi, where neat CBP layer is assumed to be EML. Both the waveguide modes (TE_0 and TM_1) are excited in the ITO OLED and experience significant energy dissipation in the entire emission wavelength, but the waveguide modes are only guided at short wavelength range in the DMD OLED with little trapped energy. Therefore, the waveguide mode suppression observed in the DMD device can yield improved EQE than the ITO counterpart. This is indeed verified in the experiment (Fig. 6). The two OLEDs have almost the same EQE when there is no outcoupling method used. However, the DMD OLED has higher portion of the energy in the substrate mode due to the waveguide mode removal in the OLED, thus shows higher EQE enhancement with the substrate mode extraction by IMF. DMD is fabricated by physical vapor deposition and can essentially be made on any type of substrate. To demonstrate its utility, an OLED is fabricated with an ultra-thin substrate, ~ 1 μm thick PVA, which becomes a foldable OLED. Fig. 6c show it is lit up when wrapped around a sharp razor blade.

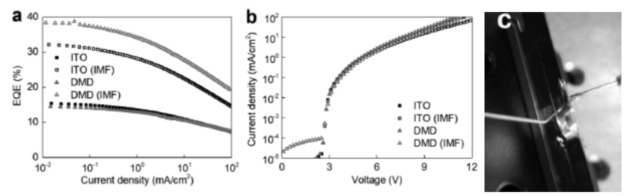

Figure 6. Performances of ITO and DMD based OLEDs. (a) EQE – J or (b) J – V relation of ITO and DMD devices, where the substrate mode is extracted by IMF.(c) An ultra-thin OLED lit up even when folded against a razor blade.

4. Expanding to tandem OLEDs

The existence of cutoff thickness for guided mode elimination when using thin Ag as electrode seems to suggest that such mechanism will disappear when the organic layers become too thick; therefore tandem OLEDs will still have to face waveguiding problems. But we will show that the tandem OLEDs can also eliminate the waveguide mode by inserting ultrathin metal layer in the charge-generation layer (CGL) layer. In theory, the waveguide mode can also be suppressed even in a tandem device as long as sufficient thickness of Ag film is inserted in the device.

As a preliminary study, we calculated waveguide mode cutoff condition in tandem OLED structures, and compare the conventional ITO based tandem OLED vs Cu-Ag based tandem OLED. We compared an ultrathin Ag based CGL with the conventional CGL of 50 nm Li doped ETL and ultrathin metal films. Figure 7a shows a calculated tandem OLED structure and Figure 7b shows effective index of fundamental waveguide mode, TE_0 mode. TM mode is not supported when TE mode is cutoff.

CGL can be 50 nm alkali doped ETL material or 1 nm Al layer for reference tandem OLED. As shown in Figure 7b, tandem OLEDs with ITO anode (reference) can excite waveguide mode along the entire organic layer thickness, meaning that the significant amount of light is trapped inside the device that lowers EQE. However, Cu-Ag based tandem OLEDs show cutoff behaviors at about 100 nm organic layer thickness. This means that Cu-Ag tandem OLED does not have trapped light and can increase device EQE in the end.

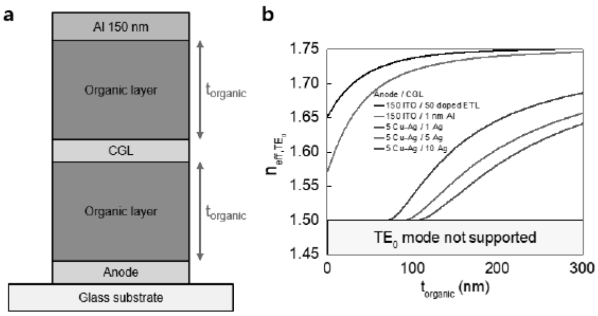

Figure 7. Waveguide mode analysis. (a) Device structure under calculation. (b) Effective index of TE_0 mode. n_{glass} = 1.5.

5. Conclusion

Coupling of the emitted light to the waveguide mode represents a significant waste of photons generated from the emission layers. Unfortunately, conventional TCO is prone to this waveguide mode as its indispensably thick layer will only serve to aid this waveguide mode. In this work, we introduced a design strategy to utilize high performance thin Ag based transparent conductor. By replacing conventional ITO with an ultrathin Cu-Ag film, the total thickness of OLEDs is maintained thinner than a cut-off thickness for the formation of an optical mode of the layers, thereby suppressing the waveguide mode in the device. This simple and yet novel approach could pave the way to replace ITO and increase efficiency in flexible and transparent OLEDs that is widely used in display industry.

6. Impact

Extraction of waveguide mode is the biggest challenge in the OLEDs community, and most of the efforts to outcouple waveguide mode has been focused on inserting patterns or grids on the ITO anode. For conventional TCOs such as ITO or IZO, to lower the resistance will need to increase the thickness of TCO but inevitably strengthens the waveguiding that traps the emitted light. This work aims to address the root cause to completely remove waveguide mode by using a sub-10 nm thick thin Ag film as anode, which offers low resistance and high transparency for practical OLED applications.

7. Acknowledgements

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8. References

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