

Extraction of surface plasmons in organic light-emitting diodes via high-index coupling

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Abstract: The efficiency of organic light-emitting diodes (OLEDs) is still limited by poor light outcoupling. In particular, the excitation of surface plasmon polaritons (SPPs) at metal-organic interfaces represents a major loss channel. By combining optical simulations and experiments on simplified luminescent thin-film structures we elaborate the conditions for the extraction of SPPs via coupling to high-index media. As a proof-of-concept, we demonstrate the possibility to extract light from wave-guided modes and surface plasmons in a top-emitting white OLED by a high-index prism.

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References and links

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1. Introduction

Organic light-emitting diodes (OLEDs) are flat and thin large-area light sources that have the potential for highly efficient, long-lived and environmentally friendly light sources. Continuous progress in science and development has led to a steady improvement of their luminous efficacy, i.e. the optical power within the spectral range visible for the human eye divided by the electrical input power. One of the main challenging problems, however, in increasing their light output is the avoidance or recovery of optical energy trapped within the OLED structure, in particular in wave-guided modes (WGMs) or surface plasmon polaritons (SPPs).

As shown schematically in Fig. 1 an OLED typically consists of organic layers sandwiched between a metallic cathode and a semitransparent anode on a glass substrate. In such a thin film structure an excited molecule can couple to different optical modes as sketched in the figure. Quantitative calculations, treating the emitting molecules as classical electrical dipoles with random orientation, reveal that in planar OLED stacks typically around 50% of the light is trapped in wave-guided and plasmon modes [1, 2], which amounts to a significant energy loss.

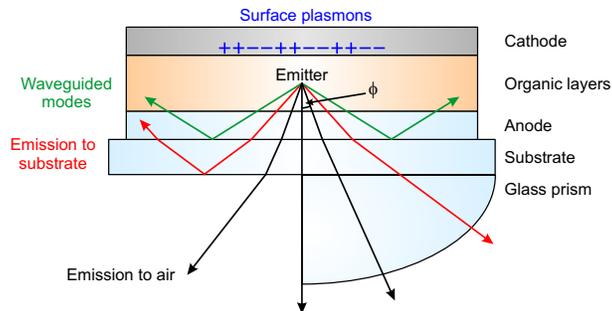


Fig. 1. Schematic illustration of an OLED with its optical loss channels: Emission to air is always present; emission to the substrate due to total internal reflection at the substrate-air-interface can be avoided by e.g. using a glass hemisphere with appropriate refractive index (cf. right part of the figure); WGMs within the organic layers and SPPs at the organic-metal-interface are hardly accessible, but account for up to 50% of energy losses.

In principle, there are three methods to reduce energy trapped in WGMs and SPPs [3]: (i) Grating coupling takes advantage of shifting the in-plane wave-vector of light by multiples of the wave-vector of a periodic grating to fulfill energy and momentum conservation of trapped modes and light propagating in air [4]. (ii) Non-isotropic dipole orientation can avoid the coupling to SPPs by appropriate molecular orientation thus favoring radiation to air [5], but is limited to special types of molecules. (iii) Index coupling will be explained in detail in the following.

Index coupling matches the refractive index of the emission layer (EML), where the light is generated, and the outside world, either by bringing the refractive index of the EML down or by using a high-index (HI) substrate. Simulations show that if the refractive index of the EML could be reduced to $n = 1$, a theoretical limit of almost 70% direct emission to air would be possible [6]. An experimental realization might, however, be difficult – simply because no

such materials are available, but lowering n_{EML} below the value of glass already would bring an enormous boost in efficiency.

The use of glass substrates with higher refractive index than the organic layers (including ITO) is possible [7–10], so that a large fraction of the light from WGMs goes into the HI glass substrate, from where it can be extracted more easily. However, due to momentum mismatch (see below for details) the energy contained in SPPs traveling at the metal-organic interface of the top electrode is not accessible in this way. Instead, as we will work out in the following, the application of a high-index medium on the back side of a thin cathode layer allows for the extraction of both SPPs and WGMs in a top-emitting OLED structure. In detail, however, additional aspects need to be considered due to the fact that the involved layers are not thick enough to be treated as bulk material. First, the evanescent field of SPPs can extend through an adjacent thin organic layer and probe an averaged refractive index and second, if the metal is thin enough, SPPs can exist on both sides of the metal layer and interact with each other [11].

2. Basic properties of surface plasmon polaritons

For the understanding of the following parts, a short introduction to SPP will be given; for profound explanations, we refer to Ref. [12, 13].

SPPs are longitudinal, p-polarized waves traveling at the interface between a metal and a dielectric with evanescent fields decaying exponentially into both adjacent media. For semi-infinite layers their dispersion relation is given by:

$$k_x = k_0 \left(\frac{\epsilon_1 \cdot \epsilon_2}{\epsilon_1 + \epsilon_2} \right)^{1/2} = \frac{\omega}{c} \left(\frac{\epsilon_1(\omega) \cdot \epsilon_2(\omega)}{\epsilon_1(\omega) + \epsilon_2(\omega)} \right)^{1/2} \quad (1)$$

with $\epsilon_{1,2}(\omega)$ being the complex dielectric functions of the metal and the adjacent dielectric layer, respectively, and k_0 the vacuum wave vector.

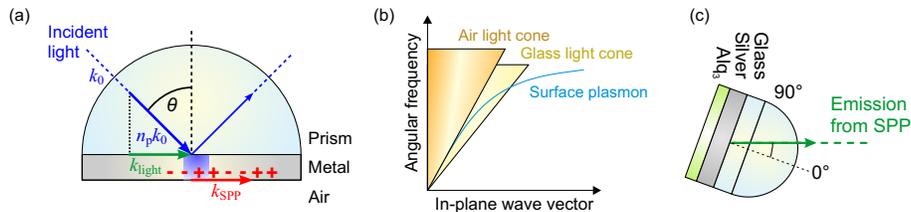


Fig. 2. (a) Kretschmann configuration: incident light with a fixed wavelength can fulfill the dispersion relation for SPP at a silver-air-interface at a specific angle. (b) Schematic SPP dispersion curve for a silver-air interface together with the light cones for far-field radiation on both sides of the metal layer. In an OLED, air as dielectric medium has to be replaced by an organic luminescent film with refractive index typically around 1.7 to 1.8. (c) Inverted Kretschmann configuration: by exciting dye molecules, SPPs at the organic-metal-interface can be excited via near-field coupling, which are then coupled out into an attached glass prism.

As an example, Fig. 2(a) shows the situation at a metal-air interface. By comparing the dispersion curves of the SPP mode and the light-line in the dielectric medium (in this case air, see Fig. 2(b)), it is obvious that they do not intersect for finite frequencies, thus energy and momentum conservation cannot be fulfilled simultaneously and as a consequence SPPs cannot couple to far-field radiation. This problem can be circumvented in the well-known Kretschmann configuration (Fig. 2(a)) [14], which makes use of the fact that the in-plane wave vector within an attached prism is stretched by a factor given by the refractive index of the prism. If the

refractive indices (and the angle) are properly chosen, it is thus possible to couple SPPs, which are evanescent modes at the metal/dielectric interface (the bottom side in Fig. 2(a)), to become radiative modes at the opposite side of the metal layer (here the top-side) and thus to extract light from SPPs in an *inverse* Kretschmann configuration (Fig. 2(c)).

3. Criteria for light extraction from SPPs

To realize this concept of index coupling in OLEDs, however, some prerequisites have to be fulfilled. First, the metal film obviously has to be semitransparent. Since there are two counteracting processes, namely an enhanced coupling of the emitting dipoles to SPPs with an increasing metal thickness on the one hand and a concomitant reduced optical transmission on the other hand, there is typically an optimum thickness, which is at around 50 nm for light emission in the green spectral region [3, 15]. More importantly, the refractive index on the top side of the metal (the "outside world") has to be larger than on the bottom side, where the OLED stack would be located, in order to match the dispersion relation of the plasmon (traveling at the bottom side) with the far-field light-line in the prism at the top side. Due to the evanescent nature of SPPs this condition not only depends on the bulk values of the organic material and the prism, but in particular on the thickness of the organic layer. Since the decay length of the SPP field amplitude perpendicular to the metal/dielectric interface is typically of the order of half a wavelength [16], the SPP field probes an extended vertical distance to the metal surface. In order to account for this feature we have introduced an *effective* refractive index n_{SPP} that has to be entered in Eq. 1 to calculate the correct SPP dispersion relation.

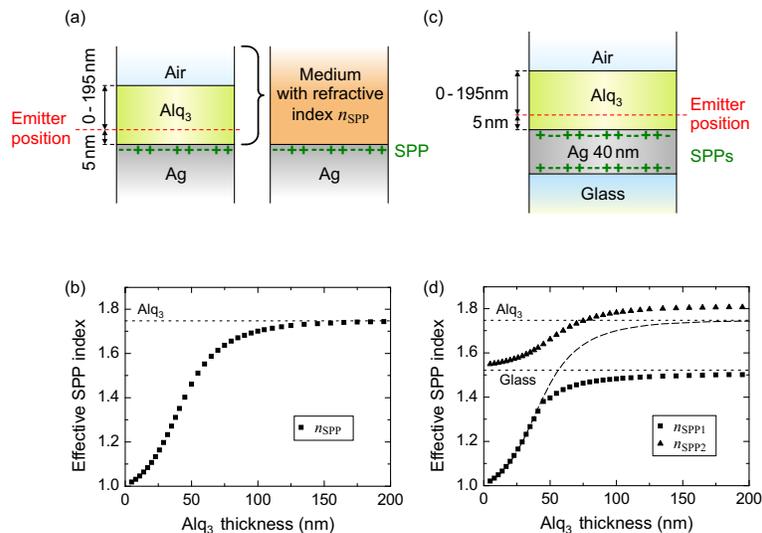


Fig. 3. Stack layout and definition of the effective SPP index: (a), (b) For simplicity the emitter position is assumed at a distance of 5 nm from a semi-infinite silver layer and the distance to semi-infinite air is varied from 0 nm to 195 nm. The *effective* SPP index is determined by assuming a semi-infinite medium with a refractive index n_{SPP} close to the silver layer so that the calculated analytical SPP dispersion (according to Eq. (1)) coincides with the numerically simulated one for a stack with finite Alq₃ layer thickness. (c), (d) The same for a finite Ag thickness of 40 nm, where two SPP modes exist in the Ag film, one at each interface.

This concept of an effective SPP index and its dependence on the thickness of the organic layer for a one-sided SPP (thick metal) and a two-sided SPP configuration (thin metal) is shown in Fig. 3 for a simplified OLED structure with a single luminescent layer (here the green fluorescent dye tris(8-hydroxyquinoline), Alq₃) in the vicinity of a silver film. As shown in Fig. 3(b), the effective index relevant for the SPP dispersion increases monotonically from the value of air ($n = 1$) to bulk Alq₃ with increasing Alq₃ thickness for the case of a semi-infinite Ag layer. This behavior changes, however, if the thickness of the Ag layer is finite (see Fig. 3(d)). For a thin Alq₃ layer the index relevant for the SPP traveling at the Alq₃/Ag interface follows the same trend as in Fig. 3(b), implying that its index is smaller than the one of the glass prism and thus energy from this SPP mode can be coupled through the thin metal film. However, at an Alq₃ thickness of about 50 nm, where the thickness dependent effective indices of the SPPs on both sides of the Ag layer would cross each other (dashed and dotted lines in Fig. 3(d)), there is actually an anti-crossing of the two curves and the assignment of their indices to SPPs at both interfaces of Ag becomes the opposite way. This means that for Alq₃ thickness larger than this critical thickness the SPP traveling at the Alq₃/Ag interface now has a larger index than glass and can no longer be extracted through the Ag layer. Consequently, for thicker organic layers, and in particular for the thickness typically used in OLEDs, the extraction of SPPs (in addition to WGMs which can also be extracted by this method) requires a medium on the back side of the metal that has a significantly higher refractive index (see below).

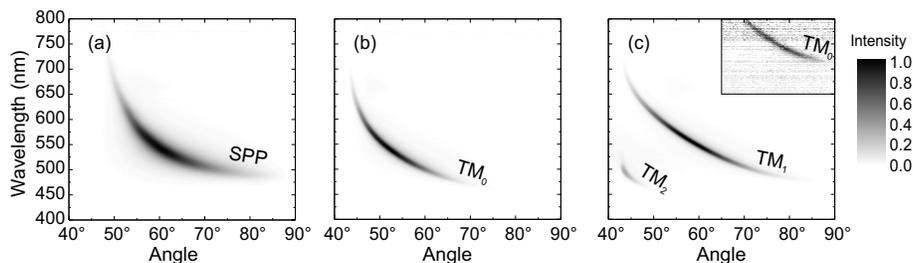


Fig. 4. Experimental out-coupled p-polarized modes of stacks with different Alq₃ thicknesses by using the inversed Kretschmann configuration (see Fig. 2(c)). (a) 40 nm Ag and 30 nm Alq₃; the Alq₃ thickness is thin enough so that the effective refractive index of the SPP is below the refractive index of the substrate and the SPP at the Ag-Alq₃-interface can be coupled out. (b) 60 nm Ag and 210 nm Alq₃; the first TM-mode (TM₀) is coupled out. (c) 50 nm Ag and 460 nm Alq₃; three modes (TM₀-TM₂) can be coupled out (the intensity within the rectangle from 65° to 90° and 650 nm to 800 nm was rescaled to make the TM₀-mode more visible). The experiments were carried out by laser-excitation of the Alq₃ (375 nm, 15 mW).

The basic experimental realization of this concept was carried out with a fused silica glass prism covered with about 50 nm Ag and variable thickness of the Alq₃ layer (cf. Fig. 4). By photoluminescence excitation of the Alq₃ molecules via laser radiation (375 nm, 15 mW) and near-field-coupling to SPPs or WGMs a part of the radiation can be extracted to the far-field. Thereby the angular dependent emission spectra show a characteristic dependence on wavelength for different modes. As can be derived from Fig. 4(a), the peak of the SPP at a wavelength of 530 nm is located at about 63° for thin Alq₃ (30 nm). For larger thicknesses of the Alq₃ layer, the SPP moves to higher angles and finally cannot be coupled out at the given wavelength, if the thickness of Alq₃ is larger than 50 nm (not shown). Increasing the thickness even further, waveguide modes within the organic layer occur, which can be detected as leaky modes with the same setup, if the Ag layer is thin enough. In principle, both transverse electric (TE) and transverse magnetic (TM) modes do exist, but for the chosen polarization of the de-

tection setup only TM polarized ones are recorded in the experiment. The experimental results with a 210 nm and a 460 nm thick Alq₃ layer are shown in Fig. 4(b) and 4(c), respectively. By comparison with simulation (see below) these modes can clearly be identified as TM₀, TM₁ and even TM₂ for the thickest Alq₃ layer.

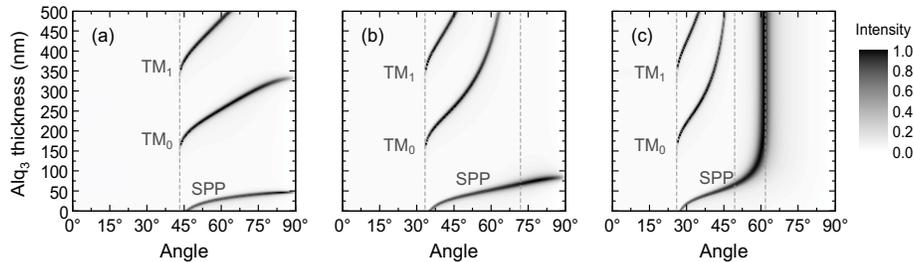


Fig. 5. Simulated angular dispersion of the p-polarized modes for (a) BK7, (b) SF6 and (c) LiNbO₃ prism attached to a 40 nm thick Ag layer and an Alq₃ film with varying thickness at a wavelength of 530 nm. WGMs are accessible for arbitrary Alq₃ thicknesses with the SF6-outcoupling structure, whereas the extraction of SPPs with Alq₃ films thicker than 100 nm requires a substrate with very high refractive index, like LiNbO₃.

For a more profound analysis of the occurrence of SPPs and WGMs, Fig. 5(a) shows exemplarily the calculated angular dispersion of p-polarized modes for a stack with 40 nm Ag and a variable Alq₃ thickness at a wavelength of 530 nm. As indicated in Fig. 3(c), for the calculations we have used the simplifying assumption that all the light is generated in an infinitely thin emission zone located 5 nm away from the Ag layer. This does certainly not reflect the true emission profile in the experiment, but is motivated on the one hand by the fact that there is non-radiative energy transfer to metal surfaces (“quenching”) in the immediate vicinity of the metal, and on the other hand by the exponentially decaying SPP field with the distance from the metal surface. In spite of these simplifications there is good agreement between the experimental data shown in Fig. 4 and the predicted mode position for glass substrates at the chosen wavelength of 530 nm. In particular, one can clearly see once again that the maximum Alq₃ thickness for a glass prism, at which the SPP can still be coupled out, is already reached at around 50 nm, which is clearly too low for commonly used OLED stacks. Using high-index glass this limitation is shifted to a higher Alq₃ thickness, e.g. for SF6 ($n = 1.82$) (Fig. 5 (b)) to 100 nm (Fig. 5 (b)). For LiNbO₃ ($n = 2.28$) (Fig. 5 (c)), the refractive index is so high that there is no limitation for the thickness any more and all modes of the organic film are accessible under an angle of not more than 60°.

4. Application to OLEDs

The application of the HI coupling concept to a white OLED with a thin semi-transparent Ag top-contact is shown in Fig. 6. The chosen OLED structure consists of a standard bottom-emitting RGB (red-green-blue) stack on an ITO-glass substrate, where the cathode is an only 30 nm thick Ag layer. The thin emission layer is located at a distance of about 65 nm away from the cathode and the whole OLED stack has a total thickness of about 100 nm.

On top of that structure a SF6 high-index glass prism was attached by index matching fluid. The photograph clearly shows that only the air mode is emitted to the bottom side, whereas all other modes (including substrate emission, WGMs and SPPs) are emitted in characteristic angular ranges to the top side. We note that the index of the used SF6 prism was not high enough to extract the SPP mode completely, so that only the red part of the SPP branch is seen under large angles close to 90°. Furthermore, the used index matching fluid was strongly absorbing in

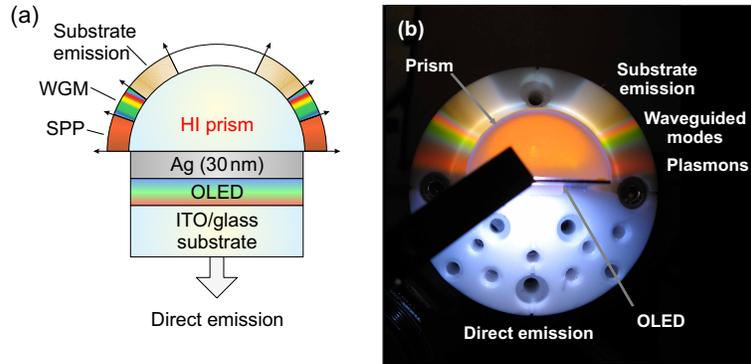


Fig. 6. White OLED with attached SF6-prism: (a) stack layout (b) photograph of angular dependent light emission into different optical modes.

the short wavelength range so that the substrate light has a brownish appearance. Nevertheless, this is a clear demonstration that the concept of high-index coupling is not only applicable to extract WGMs in bottom-emitting OLEDs, as was demonstrated before by different authors [8–10], but moreover to get access to *all trapped modes* – including the surface plasmons – in a top-emitting structure.

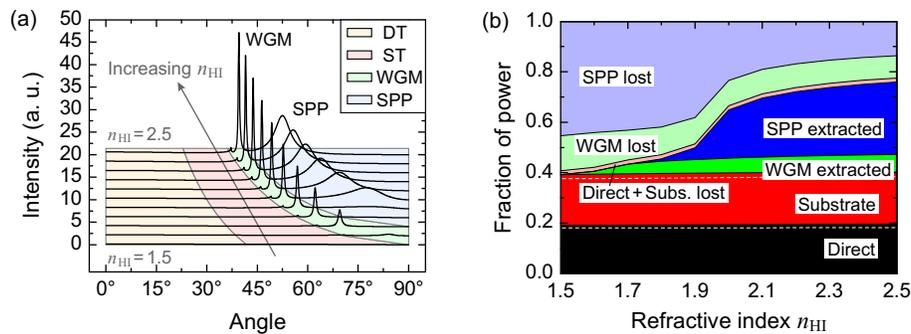


Fig. 7. (a) Simulated top emission intensities of trapped modes from Fig. 6(a) – with a semi-infinite bottom substrate – at a wavelength of 600 nm as a function of the refractive index n_{HI} of the HI glass prism on the top side. Acronyms used in the figure: direct top (DT), substrate top (ST), waveguide modes (WGM), surface plasmons (SPP). (b) Angle integrated top and bottom emission of the different extracted modes in comparison to the remaining loss-channels not accessible at the given refractive index of the prism.

In order to determine the requirements for the extraction of the SPPs and other modes by this method and to quantify the actual fraction of power that is coupled out for a given HI material, Fig. 7 shows optical simulations of light outcoupling with increasing refractive index of the HI prism for the stack given in Fig. 6(a). Note that the substrate was assumed to be semi-infinite so that no back-reflection of substrate emission to the top side occurs. Thus direct and substrate emission are negligible for the emission to the top side. On the other hand, emission from a WGM and the SPP mode emerge to the top side, shifting to lower angles and higher intensity with higher refractive index of the prism. The outcoupling of WGMs sets in at a refractive index of $n_{\text{HI}} = 1.6$, whereas the SPP mode needs a somewhat higher refractive index close to $n_{\text{HI}} = 2.0$ for gaining appreciable intensity. For a more quantitative analysis of the extraction efficiency, the angle integrated power of all modes – including the light emitted to the bottom

side – together with the amount of power trapped inside the OLED stack are plotted in Fig. 7(b). As can be clearly seen, the onset of the extraction of SPPs in fact lies at about $n_{\text{HI}} = 1.7$, but a significant boost in SPP extraction efficiency requires $n_{\text{HI}} \geq 2.0$. However, for values of n_{HI} , which are large enough, far more than 50% of the power trapped in SPPs can be extracted at the chosen wavelength, amounting to almost the same fraction as from direct and substrate emission taken together. It is also clear that the extraction of energy from WGMs will not be that efficient, because their mode profile does not overlap too much with the thin cathode layer. Nevertheless, simulations predict an overall light outcoupling efficiency of more than 70% (for the given OLED structure at a wavelength of 600 nm), thus demonstrating the great potential of this method.

5. Conclusion

Combining optical simulations and experiments on simplified thin-film structures we have been able to quantify the requirements for the extraction of SPPs at the interface between a luminescent organic film and a metal electrode via coupling to a high-index medium. Furthermore, we have demonstrated that by using an inverse Kretschmann configuration it is possible to gain access to all optical modes trapped inside a white top-emitting OLED structure. However, as shown by simulations, significant gain in efficiency can only be obtained for refractive indices of the HI medium larger than 2.0. In addition, it should be noted, that these results can only serve as a proof of principle, because using a high-index prism is not viable in practical applications. Therefore a promising way for improving the efficiency of top-emitting OLEDs will be the development of thin-film solutions with a refractive index, which is high enough, to keep the unique form factor and avoiding the viewing angle dependent color shift, e.g. by scattering elements.

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