Polaritonic OLEDs with TADF Emitters Enable Narrowband, Angle-Independent and Ultra-Efficient Emission for Brilliant Displays

Andreas Mischok*, Simon Lennartz*, Vanessa Gruber*, Francisco Tenopala-Carmona*, Julia Witt*, Sabina Hillebrandt*, Malte C. Gather*,**

*** Humboldt Centre for Nano- and Biophotonics, Department of Chemistry and Biochemistry, University of Cologne, Greinstr. 4-6, 50939 Köln, Germany**

**** SUPA, School of Physics and Astronomy, University of St Andrews, North Haugh, St Andrews KY169SS, United Kingdom**

andreas.mischok@uni-koeln.de, malte.gather@uni-koeln.de

Abstract

Thermally activated delayed fluorescence (TADF) enables highly efficient OLED emitters. We demonstrate a strategy to narrow their inherently broad emission spectrum by creating polaritons through coupling with an assistant strong-coupling layer. This reduces the emission width from 86 nm in the reference device to 24 nm, producing angle-independent brilliant green emission near the BT.2020 standard and more than doubles the efficiency over prior polaritonic OLEDs.

Author Keywords

OLED; TADF; polariton; strong coupling; display; BT.2020; angle-independent; narrowband; color brilliance.

1. Introduction

Organic light-emitting diodes (OLEDs) have revolutionized the display industry over the past few decades, establishing themselves as the preferred technology for high-end consumer devices such as smartphones, televisions, smartwatches, and more. Their success can be attributed to their high efficiency, ease of manufacturing, and chemical versatility. However, one persistent challenge for OLEDs is their relatively broad emission linewidth, which arises from inherent material disorder. In particular, highly efficient emitter molecules employing thermally activated delayed fluorescence (TADF) suffer from broad spectra, as the emitter design often relies on charge transfer between donor and acceptor moieties, leading to conformational disorder and broadband emission. This limitation poses a barrier to achieving the exceptional color purity defined by the BT.2020 standard for next-generation RGB displays, which requires narrow emission linewidths at specific emission wavelengths that are to remain unchanged across all viewing angles.

To address this challenge, resonant microcavities can be incorporated in the OLED structure. These microcavities, created by placing highly reflective, yet semi-transparent surfaces around the emitting layer, can selectively enhance specific wavelengths of light through constructive interference, effectively narrowing the emission spectrum. Such cavity designs are also particularly appealing for top-emitting OLEDs integrated with CMOS backplanes, especially for microdisplay applications.

Despite these advantages, microcavities introduce a significant drawback: their emission properties are highly sensitive to the viewing angle. As the angle increases, the resonance condition shifts, causing a noticeable and undesired blueshift in the emitted light. Recently, we have shown that this angular dependence can be addressed by harnessing strong light-matter coupling within the OLED structure [1]. In this regime, photons and excitons interact to form hybrid states known as polaritons, splitting into upper and lower energy branches (referred to as UP and LP, respectively) through Rabi splitting. By carefully tuning the coupling strength

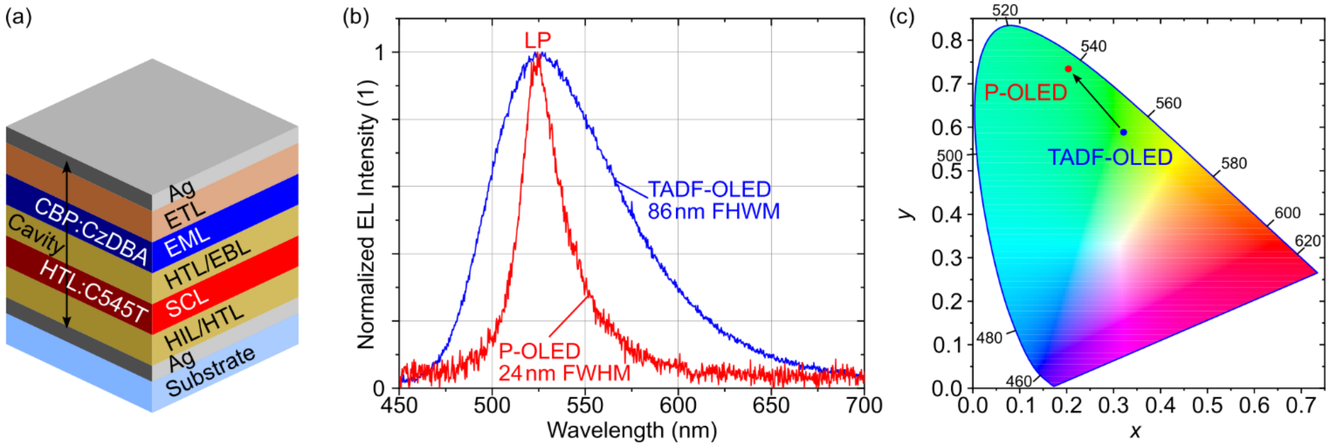

Figure 1. (a) Stack design for a TADF-P-OLED. Inserting an assistant strong coupling layer (SCL) into the hole transport layer enables polariton formation without affecting the TADF emission layer (EML)**.** (b),(c) Electroluminescence from the lower polariton state (LP) of the P-OLED is signficantly narrowed compared to the emission from a reference TADF-OLED (b), in turn the CIE coordinates of the P-OLED are shifted towards pure green emission (c).

and the offset between the resonant wavelength of the microcavity and the exciton energy, the resulting polariton dispersion can closely resemble the angle-independent excitonic dispersion, effectively minimizing angular dependence. Until recently, such polaritonic OLEDs (P-OLEDs) showed low external quantum efficiency (EQE, <1%) and low maximum luminance (<100 cd/m^2), which was mostly because achieving strong coupling in a microcavity requires the material to which the photons couple to show significant light absorption. This in turn results in a need for the emissive material to be present at high concentration, leading to concentration-quenching effects. To address this challenge, we recently demonstrated a new device architecture based on an assistant strong-coupling layer (SCL), which allowed us to realize phosphorescent P-OLEDs with EQEs around 10% and greatly improved peak luminance [1].

Here, we more than double the efficiency of P-OLEDs compared to our earlier record by translating the assistant SCL strategy to a highly efficient TADF device architecture that we further modify by introducing doped charge-transport layers for attractive operation voltage. This work demonstrates the strong potential of TADF polaritonic OLEDs (TADF-P-OLEDs) to deliver both ultra-narrow and angle-independent emission and thus makes them a promising candidate for achieving the next generation of high-performance displays.

2. Polaritonic OLEDs

The creation of polaritons requires molecules with strong absorption and small Stokes-shift, which leads to considerable challenges when trying to design highly efficient OLEDs. This is because emitter materials are usually only used in small concentrations doped into a transparent host material, limiting their total absorption. In addition, highly efficient triplet-harvesting phosphorescent and TADF-based emitters do not show strong absorption close to their peak emission wavelength. In order to solve this issue, we introduce a highly absorptive material into the hole transport layer (HTL) of a reference OLED stack. This assistant strong coupling layer (SCL) provides the necessary light-matter interaction required for polariton formation. Previously, we have shown this strategy to be effective in highly efficient phosphorescent OLEDs based on state-of-the-art Iridium-complex emitters [1]. In doing so, we were able to realize P-OLEDs with narrowband emission, negligible angular color shift and device efficiencies of up to 10% external quantum efficiency (EQE), even at high coupling strengths. By decoupling exciton recombination and light emission in the emitter from polariton formation in the SCL, we can guarantee high device efficiency while keeping the benefits of polaritonic cavities – namely the narrowband, angle-independent spectrum.

We have now realized P-OLEDs utilizing a highly efficient TADF emitter, namely 5,10-bis(4-(9H-carbazol-9-yl)-2,6-dimethylphenyl)-5,10-dihydroboranthrene (CzDBA) [2]. We carefully match CzDBA to a suitable assistant SCL, 10-(2-Benzothiazolyl)-2,3,6,7-tetrahydro-1,1,7,7-tetramethyl-1H,5H,-11H-(1)benzopyropyrano(6,7-8-I,j)quinolizin-11-one (C545T), and sandwich the entire device stack in a second-order microcavity (Figure 1a). To do so, we first introduce a semitransparent silver mirror as anode paired with an opaque aluminum cathode to form the microcavity. Tuning the cavity resonance in TADF-OLEDs proves more challenging than for established phosphorescent architectures, as device efficiency depends critically on the charge balance of holes and electrons in the emission layer. In order to achieve the necessary cavity thickness, we furthermore include an electrically doped hole transport layer into an optimized reference device. The high conductivity of the doped HTL allowed us to increase the cavity thickness without affecting charge balance.

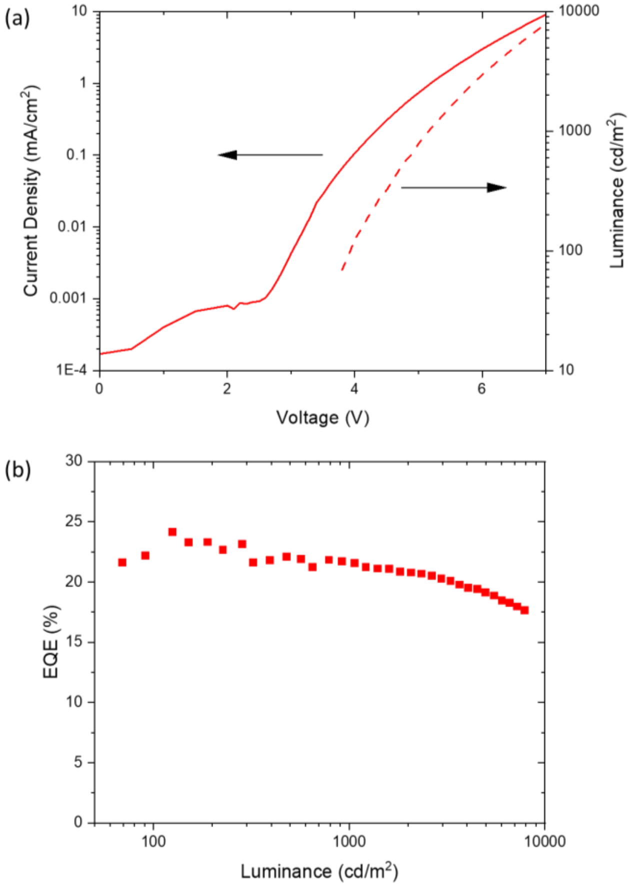

Figure 2. (a) jVL characteristics of TADF-P-OLED and (b) EQE as function of device luminance.

We also made reference devices based on the same emitter, following the optimized CzDBA device stack reported in [2]. When comparing the electroluminescence (EL) spectra of the two devices, we find that the P-OLED shows a drastically narrowed emission spectrum with a full-width-half-maximum (FWHM) of 24 nm compared to 86 nm in the reference device (Figure 1b). Due to careful tuning of the cavity thickness, the peak emission wavelength of CzDBA is retained in the P-OLED device, with an EL maximum at a wavelength of approximately 525 nm.

The spectral narrowing and removal of the long wavelength tail from the EL spectrum leads to a marked shift in CIE coordinates, toward a more saturated green color point (Figure 1c). This illustrates the potential of our strategy to use literature-known TADF emitter materials with prohibitively broad intrinsic emission spectra in future BT.2020 compliant displays.

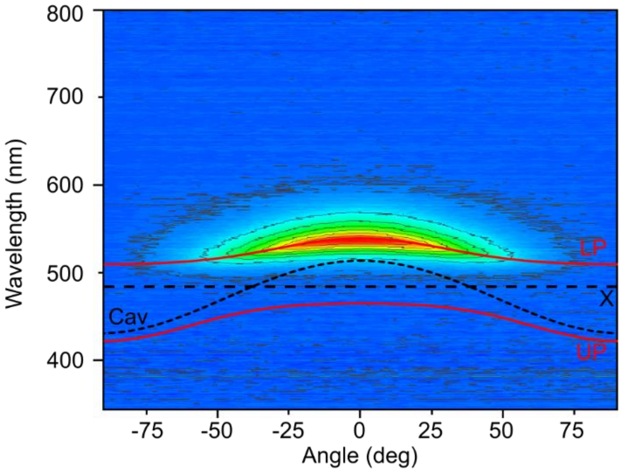

Figure 3. Angle-resolved electroluminescence spectrum of the TADF-P-OLED**.** The parabolic microcavity resonance (Cav) and angle-independent exciton resonance (X) can strongly couple, forming hybrid lower and upper polaritons (LP,UP) with reduced angular dependence. As the device emission follows the LP resonance dispersion, the angular shift is reduced to 15 nm over the full angle range.

Our TADF-P-OLEDs show a peak EQE of 24% with minimal roll-off to 21.5% at 1000 cd/m^2. The devices generate a luminance of 1000 cd/m^2 at 5.2 V and reach nearly 8000 cd/m2 at 7 V (Figure 2).

Conventional microcavity OLEDs with narrow EL spectrum show a significant spectral shift with increasing viewing angle, which can lead to very noticeable changes in emission color when the device is tilted. By contrast, our P-OLED design largely prevents this effect. Figure 3 shows the measured EL spectrum over the entire forward half sphere, i.e. from -85° to + 85° in a false color representation. There is hardly any change in peak emission wavelength with angle. For comparison, the figure also shows the expected change in peak EL wavelength for a conventional microcavity OLED (Cav in Figure 3) and the predict angle dependence of the emissive LP and the non-emissive UP branch and the wavelength of the excitons in the C545T-based assistant SCL (LP, UP and X, respectively in Figure 3). This again shows the pronounced difference between the significant angle sensitivity of the conventional cavity and the near angle-independent behavior of the P-OLED.

3. Conclusion

In conclusion, we report the first efficient polaritonic OLED using a TADF emitter. Compared to all earlier reports on polaritonic OLEDs, device EQE was more than doubled, from the previous record of 10% to 21.5% at 1000 cd/m^2 in the current work. While efficiency still falls short of the current record for conventional TADF OLEDs, the TADF-P-OLED reported here provides exceptionally narrow-band, saturated emission, especially considering the very broad intrinsic emission spectrum of the CzDBA emitter that was used.

4. Impact of Your Research

Micro-cavity OLEDs are of great interest to the display industry as they are simpler to manufacture, offer better mechanical flexibility than alternative designs based on transparent conductive oxides, and can be readily integrated with a wide variety of backplane technologies, including CMOS for OLED microdisplays. However, in current architectures, one generally has to minimize the reflectance of the electrode through which light emission occurs. This is because in conventional microcavity OLEDs with more reflective electrodes, dispersion will lead to unacceptable shifts in perceived color with viewing angle.

Polaritonic OLEDs facilitate the use of microcavities with more highly reflective electrodes while at the same time improving angular color stability and color saturation. Using more reflective electrodes has the additional benefit of simplifying the deposition process (nano-island formation in the metal layer is less likely to occur) and increasing electrical conductivity.

By demonstrating that the polaritonic OLED concept can be successfully applied for TADF emitters and by breaking the important EQE threshold of 20%, our current work demonstrates for the first time that polaritonic OLEDs have a practical potential for use in future OLED displays. As their device architecture is nearly identical to existing OLED stacks, with only a single additional material required compared to existing TADF architectures, we expect that implementation in the display industry would be relatively straight-forward.

5. Acknowledgements

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6. References

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