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Transparent and color-tunable organic light-emitting diodes with highly balanced emission to both sides

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ABSTRACT

Future lighting applications will strongly benefit from transparent luminescent devices. Here, we demonstrate transparent organic light-emitting diodes (OLEDs), which provide real-time adjustment of the emission color. Making use of the AC/DC concept, two stacked subunits can be addressed independently via an AC signal. Combining blue and yellow emission leads to the possibility to tune the emitted color between deep blue over cold white and warm white to yellow on both emission sides. For such highly complex device architectures, the thickness of each layer needs to be adjusted carefully in order to achieve balanced and efficient emission in both directions. Therefore, optical simulations are carried out to optimize the OLED. Based on these simulations, we present transparent, indium-free OLEDs that achieve a luminous efficacy of 8.7 lm/W in bottom direction and 9.7 lm/W in top direction at a brightness level of 1000 cd/m^2 for warm white emission and a peak transmission of 56%. Using an emitter combination providing red, green, and blue emission, we were able to achieve a high color-rendering index (CRI) of 84, which further expands the range of possible applications for this promising device concept.

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Within the wide field of organic light-emitting diodes (OLEDs), white light emission is of particular interest, since it is an essential ability for many lighting applications. Therefore, various types of white OLEDs have been presented so far in literature [1–5]. A specific way to achieve white light emission are color-tunable OLEDs, which allow flexible adjustment of the emission color. One of the first publications on this topic showed a voltage-dependent color emission, which struggled with the fact that the emitted brightness also scaled with the driving voltage [6]. Another approach consists of two differently colored devices side by side [7]. However, this suffers from the need of additional structuring during sample preparation, which potentially increases the costs of such a system. Since many organic materials show a high transparency, vertical stacking of two or more OLEDs presents a viable alternative to pixelated concepts. Color-tunable stacked device were already presented in 1996 [8], but showed very low external quantum efficiencies (EQE) of less than 1%. Only recently, efficient color-tunable bottom-emitting OLEDs were presented [9], which are driven by an alternating current (AC) signal, achieving 36.8 lm/W at a brightness of 1000 cd/m^2. In these so-called AC/DC-OLEDs, two OLED subunits are stacked on top of each other, where the anode of one unit is connected to the cathode of the other. Thus, both subunits can be addressed independently requiring only two electrodes. This concept can be adopted for transparent applications as for example needed for window lighting applications.

Here, we demonstrate transparent, two-unit, color-tunable AC/DC-OLEDs with a transparency of 56%. This value is comparable to monochrome transparent OLEDs [10], despite the nominal doubling of OLED units. Furthermore, our devices are free of indium-tin-oxide (ITO) and thus require no potentially harmful sputtering-process. The presented devices allow warm-white light emission both in bottom-emission (BE) and in top-emission (TE). The luminous efficiency reaches 8.7 lm/W and 9.7 lm/W at a brightness of 1000 cd/m^2, respectively. Moreover, the emission color can be continuously tuned from deep blue over cold- and warm-white to saturated yellow. Using a three-color emitter combination containing red, green, and blue emission, a color-rendering index (CRI) of 84 can be achieved. So far, transparent OLEDs tended to show very unbalanced emission between top and bottom emission direction [10,11]. Contrary to this, we were able to

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achieve equalized light emission, both in terms of efficiencies and in terms of spectra.

Fig. 1a) shows the structure of the investigated OLEDs, including the thickness of each layer. On a glass substrate (Corning Eagle XG, Thin Film Devices, Inc.), a 2 nm thin layer of molybdenum-oxide (MoO_3) ensures a smooth growth of the following ultra-thin wetting layer metal electrode [2,12,13]. Two nanometer of gold lead to a Frank-van der Merwe growth of the subsequent eight nanometer thick silver layer, simultaneously providing good conductivity and transparency. On top of this electrode (E1) there are two OLED-subunits in pin-architecture, separated by another ultra-thin Au/Ag-electrode (E2). Both units are similar in their structure and contain the following materials: Spiro-TTB doped with 4 wt% F6-TCNNQ as hole transport layer (HTL). BPhen doped with cesium as electron transport layer (ETL). BPhen as hole blocking layer (HBL). Spiro-TAD as electron blocking layer (EBL). The emission layers are 4P-NPD (blue emission) and a double layer of TBPi and TCTA doped with 8 wt% $Ir(dhfpy)_2(acac)$ (yellow emission), respectively. On top, the device is covered by a third ultra-thin electrode (E3) and an organic capping layer made of α-NPD for enhanced out-coupling [10]. Chemical names and further details to the organic materials can be found in literature [1,9]. All layers are processed in vacuum (10^{-7} mbar) by thermal evaporation (Kurt J. Lesker & Co.)

Electrical measurements (current density j, voltage V) are done with a source measure unit (SMU2400, Keithley Instruments). During the measurement, the luminance (L) is recorded by a silicon photodiode. A calibrated spectrometer (CAS140, Instrument Systems GmbH) quantifies the spectral radiance in forward direction. All efficiencies are evaluated primarily under Lambertian assumption but have been corrected, measuring the angular dependency of the emission with a custom built goniometer and a calibrated spectrometer (USB4000, Ocean Optics, Inc.). The luminous efficiency of white light emission is measured under AC driving, using a digital power-meter (WT1600, Yokogawa Electric Corp.) and chroma-meter (CS-100, Konica Minolta, Inc.). An arbitrary waveform generator (33220A, Agilent Technologies, Inc.) provides an AC-signal at a frequency of 50 Hz. Measurements of the shape of the signal can be found in the supplementary (Fig. S1).

For angular stable emission spectra as well as for high efficiency, it is indispensable to place the emission layer in a maximum of the intensity of the electric field ($\vec{E}$) inside the OLED. The latter is obtained from the solution of the Helmholtz equation in media (relative permittivity ε_r and relative permeability $\mu_r = 1$):

$$\vec{\nabla}^2 \vec{E} + \omega \mu_0 \varepsilon_0 \varepsilon_r \vec{E} = 0, \tag{1}$$

where ω is the angular frequency, ε_0 is the vacuum permittivity and μ_0 refers to the vacuum permeability. The solution has the form of a plane wave $\vec{E} = \vec{E}_0 e^{i\vec{k}\vec{r}}$, with the wavevector $\left|\vec{k}\right|^2 = k^2 = \frac{\omega^2}{c^2}\varepsilon_r$. A more detailed evaluation shows that this plane wave can be decomposed into four contributions. Two are given due to $k = \pm\sqrt{\vec{k}^2}$, which can be interpreted as an upwards and a downwards traveling plane wave. Additionally, both of these terms have a contribution for s- and p-polarization [14]. The propagation of the resulting plane wave in stratified organic devices is described by the transfer-matrix formalism [14,15]. The absolute square $\left|\vec{E}\right|^2$ gives the spatial distribution of the intensity of the electric field and is fixed by setting the initial amplitude at $x_0 = 0$. In non-transparent devices the propagation direction, and thus x_0, is confined through the boundary conditions that the field-amplitude must not increase exponentially in thick metal layers. However, in transparent devices no such boundary condition exists and thus, both directions have to be taken into account. Since matrix multiplication is not commutative, this gives two independent solutions, which can be interpreted as two optical modes. Hence, a stack architecture has to be found, where the maxima for both emission directions coincide. Moreover, as $\vec{E} = \vec{E}(\vec{k})$, the intensity distribution of the electric field also depends on the wavelength. Altogether this means, that for the presented transparent two color

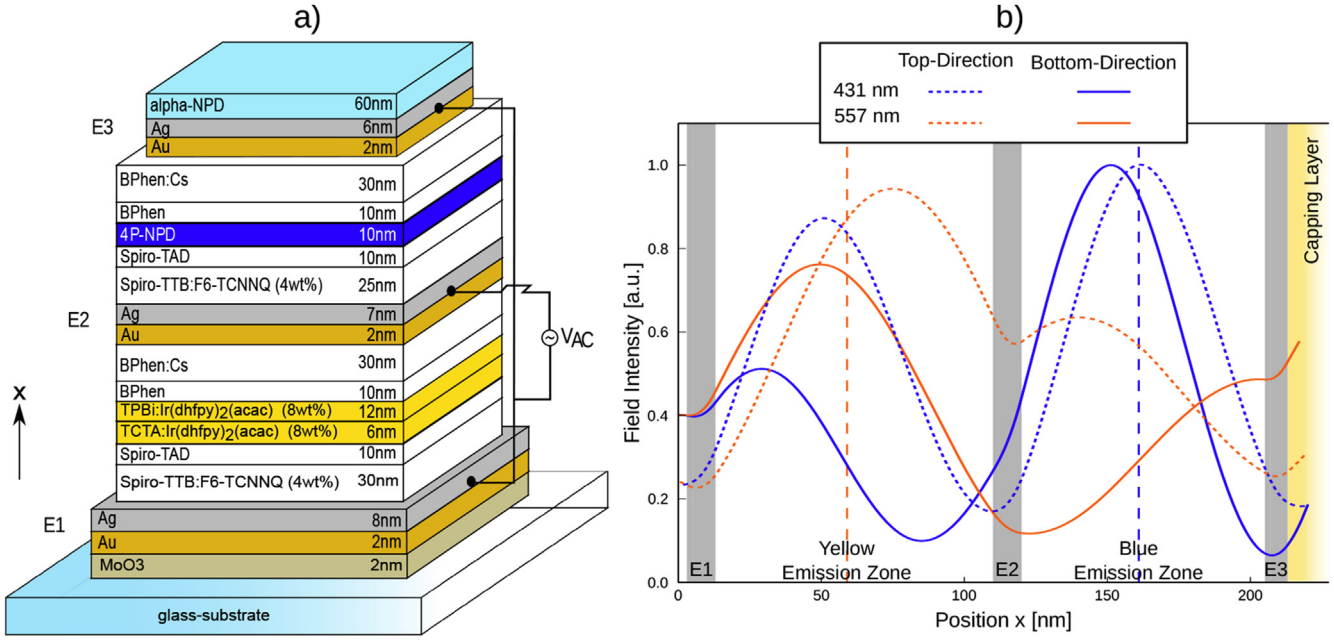

Fig. 1. a) Schematic stack architecture with layer thicknesses and contacting of the AC/DC OLED. b) Distribution of the electric field in the device. 431 nm and 557 nm refer to the PL-peak intensity of the blue and yellow emitter, respectively. The emission zones are located in the field maximum for both colors and emission directions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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