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Phosphorescent top-emitting organic light-emitting devices with improved light outcoupling

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A dielectric capping layer has been used to increase the light output and to tune the spectral characteristics of top-emitting, phosphorescent organic light-emitting devices (OLEDs). By controlling the thickness of the dielectric layer deposited on top of a thin metal cathode, the transmittance of the top electrode can be adjusted. Maximum light output is not achieved at highest cathode transmittance, indicating that the interplay between different interference effects can be controlled by means of the capping-layer thickness. Furthermore, we demonstrate that the electrical device characteristic is not influenced by the capping layer. The strength of our concept in particular lies in the fact that the optical and the electrical device performance can be optimized separately. Using the capping-layer concept, we have achieved an OLED efficiency of 64 cd/A with pure green emission. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1537052]

In recent years, major breakthroughs have led to significant improvements in the performance of organic light-emitting devices (OLEDs).[1–4] Meanwhile, the internal quantum efficiency of optimized OLEDs is gradually approaching the theoretical limits. Therefore, further improvements of the external light emission can only be expected by increasing the outcoupling efficiency.[5,6] More recently the optical design has received a great deal of attention as a means of tailoring the emission properties of OLEDs. It has been found that the external quantum efficiency and also the spectral characteristic significantly depend on the OLED architecture, in particular on the layer thicknesses, as a consequence of optical interference effects.[7–9] Fukuda and co-workers demonstrated that a change of the thickness of the hole-transporting layer (HTL) and also of the transparent indium tin oxide anode in bottom-emitting OLEDs leads to a shift of the electroluminescence (EL) spectrum and a variation of the EL intensity by a factor of 2 in the material set used.[8] The influence of the electron-transport layer (ETL) thickness on the optical properties of bottom-emitting devices was studied by So and co-workers.[7] They explained their results by wide-angle interference originating from the superposition of the amplitudes of direct emission and emission reflected from the cathode mirror. In top-emitting devices, where the EL is outcoupled through a semitransparent metal cathode as used in this report, the influence of interference effects is even stronger. In this case, not only wide-angle but also multiple-beam interference has to be taken into account.[10] Consequently, the strength of the optical interference effects depends critically on the reflectivity of the metal cathode. One concept, known from the optics of metal coatings, utilizes a thin dielectric layer on top of a thin metal film to enhance or decrease the transmittance of the cathode.[11] This approach was recently applied to top-emitting OLEDs by Hung and co-workers.[12] They demonstrated that by using an organic layer on top of a thin metal cathode a significant improvement in light output can be achieved because of the enhancement of optical transmission, and concluded that optimum performance is achieved at the highest cathode transparency.

In this letter, we report on top-emitting, phosphorescent OLEDs that exploit a dielectric layer on top of a thin metal cathode to improve light outcoupling. The wide-band-gap semiconductor ZnSe with a refractive index of $n = 2.6$ was used as capping material. Detailed investigations of the influence of the capping-layer thickness on the $I-V$ and EL$-V$ characteristics, the efficiency and the spectral characteristics are presented. It is demonstrated that the light emission is not maximum at highest transmittance of the cathode but is determined by an interplay between the different interference effects, which are governed by the thickness of the capping layer.

The devices are built on glass substrates (Schott AF45) precoated with an opaque, high-work function metal such as Pt, Ir, Ni, Pd, or Mo (≈ 75 nm thickness) as hole-injecting anode. The organic multilayer structure consists of copper phthalocyanine (CuPc) as buffer layer, N,N′-di(naphthalene-1-yl)-N,N′-diphenyl-benzidine (NPB) as the HTL, 4,4′-N,N′-dicarbazole-1,1′-biphenyl (CBP) doped with 6% tris(2-phenylpyridine)iridium [$\mathrm{Ir(ppy)_3}$] as emission layer, 2,9-dimethyl-4,7-diphenyl 1,10-phenanthrolin (BCP) as the hole-blocking layer and tris(8-hydroxyquinolinato)aluminum ($\mathrm{Alq_3}$) as the ETL. The organic layer thicknesses were optimized with the combinatorial method to be 20 nm CuPc, 40 nm NPB, 20 nm CBP, 10 nm BCP, and 40 nm $\mathrm{Alq_3}$.[9,13] In this top-emitting device architecture, the EL is observed through a semitransparent metal cathode consisting of 12 nm Ca and 12 nm Mg. The multilayer structure was further capped with a ZnSe layer to adjust the transmittance of the metal cathode. The active area of our devices was $2 \times 3\ \mathrm{mm}^2$.

The organic materials were purified by vacuum sublimation. Depositions were carried out in a high vacuum system

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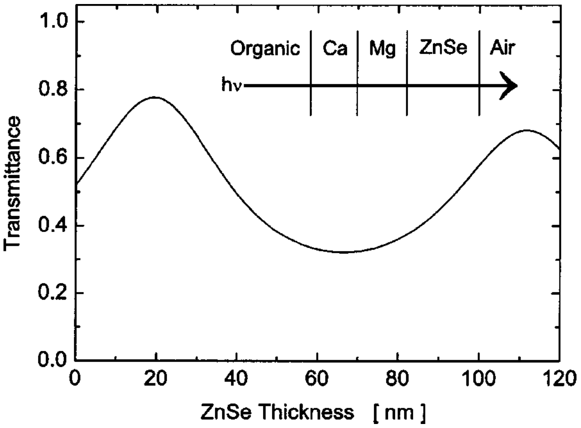

FIG. 1. Calculated transmittance of the layer stack shown in the inset as a function of ZnSe thickness. A transfer matrix method is used for the calculation, which assumes that an electromagnetic wave (510 nm) impinges from the left side, and the organic is assumed to be infinitely thick with a refractive index of $n=1.8$.

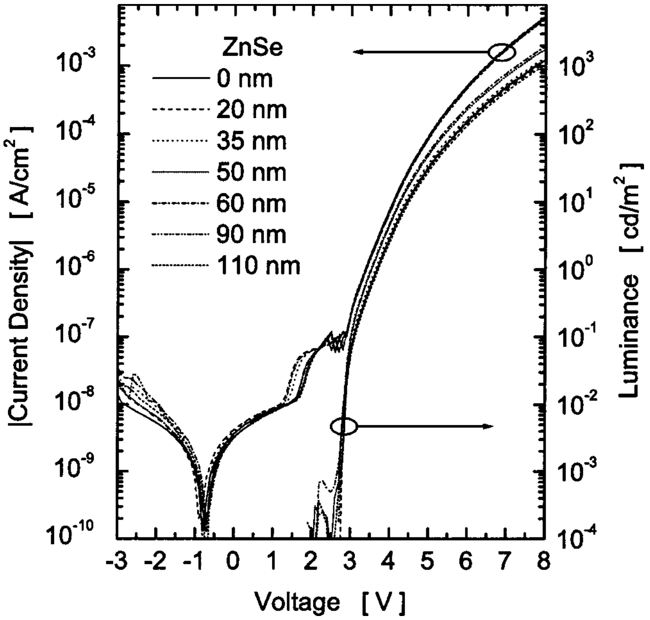

FIG. 2. Comparison of the $I-V$ and EL$-V$ characteristics measured on the same OLED after subsequent ZnSe deposition steps. The total thickness of ZnSe is shown in the figure. OLED structure: metal anode / 20 nm CuPc / 40 nm NPB / 20 nm CBP+Ir(ppy)$_3$ / 10 nm BCP / 40 nm Alq$_3$ / 12 nm Ca / 12 nm Mg / X nm ZnSe.

at a chamber base pressure ranging between 4×10^{-7} and 1×10^{-6} mbar by thermal evaporation from resistively heated boats. Typical deposition rates for the organic compounds, the metal and ZnSe were ≈1 Å/s and accordingly 0.06 Å/s for the dopant. Calibrated quartz-crystal monitors were used to individually control the deposition rates. Characterization of the OLED was performed under inert conditions in a glove-box system filled with Ar and directly connected to the deposition chamber. $I-V$ and EL$-V$ characteristics were measured with a Hewlett Packard parameter analyzer (HP 4145) and a sensitive Si photodiode (Hamamatsu S2281). The spectral characterization and the luminance calibration of the photodiode were performed with a Photo Research PR704 spectroradiometer.

Figure 1 depicts the transmittance of the Ca/Mg cathode as a function of the ZnSe capping-layer thickness calculated for a wavelength of 510 nm using a transfer matrix method.[14] The validity of the optical model used here was verified by comparing various calculated and experimental results. The calculation indicates that the transmittance of the uncovered thin Ca/Mg layer is ≈0.52 which is in agreement with experimental results. The transmittance of the Ca/Mg/ZnSe film shows an oscillatory dependence on the ZnSe thickness. With increasing capping-layer thickness, the transmittance of the cathode stack is enhanced and reaches a maximum of 0.78 at ≈20 nm ZnSe. A first minimum in transmittance of 0.32 is obtained at 65 nm ZnSe.

To study the influence of the thickness of the dielectric capping layer on the OLED performance, ZnSe was deposited on top of the Ca/Mg cathode of a phosphorescent OLED. The ZnSe layer was sequentially increased from 0 to 110 nm on the same OLED, and electrical and optical measurements were performed under inert conditions after each growth sequence. The ZnSe thickness range chosen guaranteed that the minimum and maximum transmittance of the Ca/Mg/ZnSe layer combination was covered (see Fig. 1). In Fig. 2 selected $I-V$ and EL$-V$ characteristics of the OLED described with different ZnSe thicknesses are shown. This plot clearly confirms that the evaporated ZnSe layer does not alter the electrical characteristics of our top-emitting OLED, i.e., the various $I-V$ curves and also the EL onset voltages of the OLED examined are almost identical. However, a significant influence of the ZnSe thickness on the outcoupled EL intensity of the OLED can be observed. For example, without capping layer, a luminance of 1.2×10^{3} cd/m^2 was measured in forward direction at 8 V, whereas with 60 nm ZnSe the EL intensity was increased to a maximum value of 2.0 $\times10^{3}$ cd/m^2. The dependence of the outcoupled EL intensity on the ZnSe thickness is also reflected in the behavior of the external efficiency (cd/A), which is shown in Fig. 3. Without capping layer the phosphorescent top-emitting OLED already obtains an efficiency of 38 cd/A at 4 V. With increasing ZnSe thickness the external efficiency (cd/A) first decreases slightly to 36 cd/A at 20 nm ZnSe, but increases for higher thicknesses, reaching a maximum value of 64 cd/A at 60 nm ZnSe. For even thicker dielectric layers, the external efficiency decreases again.

The fact that the $I-V$ characteristics and also the func-

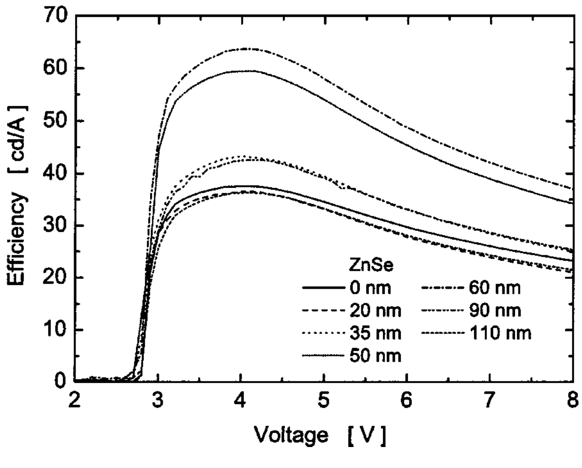

FIG. 3. Efficiency (cd/A) vs voltage of the OLED shown in Fig. 2 for various capping-layer thicknesses.

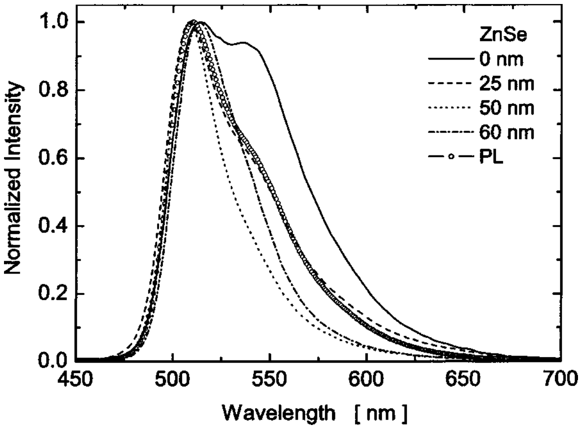

FIG. 4. EL spectra (normalized) measured at constant current density (1.67 mA/cm^2) for various ZnSe thicknesses, which were deposited on top of the same OLED as shown in Fig. 2. The PL spectrum of Ir(ppy)$_3$ doped with 6% in CBP measured using a solid film on quartz is shown for comparison.

tional behavior of the efficiency-versus-voltage curves are not influenced by the deposition of the dielectric material indicates that the electrical properties as well as the charge balance factor of the OLED are unaffected by the capping layer. These results clearly manifest that the enhancement of the external efficiency can be attributed exclusively to the modified optical architecture. The number of internally generated excitons is identical, but the number of photons detected externally is significantly affected by the modification of the optical structure.

By taking advantage of the ZnSe layer, the outcoupled EL intensity in forward direction was significantly enhanced by a factor of 1.7. The required thickness of 60 nm ZnSe for the maximum external efficiency measured, however, does not agree with the value of 20 nm calculated for the highest transmittance of the cathode configuration, but is closer to the minimum transmittance calculated (see Fig. 1). This discrepancy substantiates the fact that the simple assumption that maximum outcoupling is obtained at maximum transmittance of the cathode is not valid. The correlation between both parameters is considerably more complex. To explain the experimental results qualitatively as well as quantitatively, the interference effects present in this top-emitting structures have to be taken into consideration.[15] In the device structure considered, the wide-angle interference depends only on the reflectivity of the anode and is independent of the transmittance of the cathode used. Therefore, it is unaffected by the capping layer. In contrast, the multiple-beam interference is intensified with increasing reflectivity, and is thus strongly dependent on the thickness of the dielectric capping layer. Naturally, the capping affects the angular emission characteristics, and a detailed study will be reported elsewhere.

The interference effects and therefore the capping layer also influence the spectral characteristics. In Fig. 4 it is demonstrated that the EL spectra strongly depend on the ZnSe thickness deposited. Without ZnSe, the EL intensity is maximum at 512 nm and possesses a full width half maximum (FWHM) of 72 nm. A comparison with the photoluminescence (PL) spectrum of the Ir(ppy)$_3$ complex doped with 6% in CBP reveals that the emitted EL spectrum of the uncapped OLED is already modified by the weak cavity of the OLED structure. Owing to interference effects, the corresponding EL spectrum is broader and slightly shifted to longer wavelengths. The best agreement between EL and the PL spectra is found for 25 nm ZnSe, where the cathode transmittance is very close to maximum. A very pure green emission with a peak at 508 nm and an extremely narrow FWHM of only 36 nm resulting in 1931 CIE color coordinates of $x = 0.19$ and $y = 0.67$ is observed at 50 nm ZnSe.

In conclusion, we have demonstrated that in top-emitting devices the concept of dielectric capping is a powerful tool to improve OLED performance. The complex interplay between wide-angle and multiple-beam interference can be controlled via the thickness of the dielectric capping layer on top of the cathode. These interference effects can be exploited to tune the spectral characteristics and to improve light outcoupling. Hence, the highest light emission is not achieved at maximum transmittance of the cathode. By using this approach we were able to enhance the outcoupled light intensity in forward direction by a factor of 1.7, yielding a maximum efficiency of 64 cd/A with pure green emission.

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[1] C. W. Tang, S. A. Van Slyke, and C. H. Chen, J. Appl. Phys. **65**, 3610 (1989).
[2] J. Kido and T. Matsumoto, Appl. Phys. Lett. **73**, 2866 (1998).
[3] M. A. Baldo, S. Lamansky, P. E. Burrows, M. E. Thompson, and S. R. Forrest, Appl. Phys. Lett. **75**, 4 (1999).
[4] X. Zhou, M. Pfeiffer, J. Blochwitz, A. Werner, A. Nollau, T. Fritz, and K. Leo, Appl. Phys. Lett. **78**, 410 (2001).
[5] C. F. Madigan, M. H. Lu, and J. C. Sturm, Appl. Phys. Lett. **76**, 1650 (2000).
[6] T. Tsutsui, M. Yahiro, H. Yokogawa, K. Kawano, and M. Yokoyama, Adv. Mater. **13**, 1149 (2001).
[7] S. K. So, W. K. Choi, L. M. Leung, and K. Neyts, Appl. Phys. Lett. **74**, 1939 (1999).
[8] Y. Fukuda, T. Watanabe, T. Wakimoto, S. Miyaguchi, and M. Tsuchida, Synth. Met. **111–112**, 1 (2000).
[9] T. A. Beierlein, H.-P. Ott, H. Hofmann, H. Riel, B. Ruhstaller, B. Crone, S. Karg, and W. Rieß, Proc. SPIE **4464**, 178 (2002).
[10] K. Neyts, P. De Visschere, D. K. Fork, and G. B. Anderson, J. Opt. Soc. Am. B **17**, 114 (2000).
[11] A. Vasicek, *Optics of Thin Films* (North-Holland, Amsterdam, 1960).
[12] L. S. Hung, C. W. Tang, M. G. Mason, P. Raychaudhuri, and J. Madathil, Appl. Phys. Lett. **78**, 544 (2001).
[13] H. Riel, T. Beierlein, S. Karg, and W. Rieß, *Proc. 11th Int'l Workshop on Inorganic and Organic Electroluminescence—EL2002, Ghent, Belgium, September 2002* (Academia, Ghent, Belgium, 2002), p. 317.
[14] J. Lekner, *Theory of Reflection of Electromagnetic and Particle Waves* (Martinus Nijhoff, Dordrecht, 1987).
[15] H. Riel, Ph.D. thesis, University of Bayreuth, Germany, 2002, Chap. 6.