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Electrophosphorescent p-i-n Organic Light-Emitting Devices for Very-High-Efficiency Flat-Panel Displays**

By Martin Pfeiffer,* Stephen R. Forrest,* Karl Leo, and Mark E. Thompson

Displays are ubiquitous in our everyday world, providing the primary interface with computers, appliances, and test instruments. Some critical properties of all displays used in mobile applications are mechanical robustness, reliability, full color, light weight, and high efficiency to extend battery life. Current candidates for these advanced, mobile displays are based on back-lit liquid crystals, field-emission displays, and organic light-emitting devices (OLEDs), based on either polymers or small molecular weight materials. Until recently, the highest luminance efficiencies of devices based on these technologies have been in the range of 4–6 lm W⁻¹ at display operating brightnesses of ~100–300 cd m⁻². This situation improved dramatically, achieving green-emitting efficiencies exceeding 15 lm W⁻¹ with the demonstration of electrophosphorescent OLEDs (PHOLEDs) based on small molecular weight organic materials.^[1–4] Still, the efficiency of small molecular weight materials is limited by the voltage drop across

the highly resistive semiconducting layers comprising the devices. Thus, as is apparent in Table 1, the voltage drop across an OLED is considerably higher than the minimum potential (V_λ) needed to generate a photon of wavelength λ . Recently, however, it has been shown^[5–9] that increasing the conductivity of organic semiconductor layers by doping with either donors (for electron conducting materials) or acceptors (for hole-transport materials) can significantly reduce the voltage drop across organic thin films, and in particular, fluorescent OLEDs. This approach has recently led to the demonstration of a fluorescent p-i-n type OLED with an operating voltage of 2.55 V for 100 cd m⁻², i.e., close to the minimum drive voltage limit of V_λ .^[9] In this work, we extend this approach to small molecular weight electrophosphorescent light-emitting devices, thereby achieving very high efficiency OLEDs that perform in a manner similar to LEDs employing conventional semiconductor materials. Indeed, we obtain significant light emission at applied voltages $V \leq V_\lambda$. This advance represents a significant step in realizing very high brightness displays with efficiencies unmatched by any current competing technology. Our results also suggest that white-emitting PHOLEDs will have applications in very high efficiency room lighting in the future, as well as in high current devices such as bipolar transistors and lasers.

The doped PHOLEDs use a p-i-n heterostructure configuration, as shown in the proposed equilibrium energy level diagram in the inset of Figure 1. Here the 50 nm thick 4,4',4''-tris-(3-methylphenylphenylamino)triphenylamine (m-MTDATA) hole-transport layer (HTL) doped with 2 mol.-% of 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F₄-TCNQ) is deposited on an indium tin oxide (ITO) coated glass substrate. The dopant, F₄-TCNQ, has previously been shown to be an effective acceptor, whose electron affinity^[5,10] approximately coincides with that of m-MTDATA,^[11,12] thereby providing a high density (~10¹⁷ cm⁻³) of holes to the HTL.^[13,14] To prevent excitons generated in the emission layer (EML) from drifting into the heavily doped and highly conducting HTL where non-radiative quenching might occur, an electron blocking buffer layer of 10 nm thick tris(phenylpyrazole)iridium (Ir(ppz)₃) is deposited, followed by a emission layer of 5 nm thick N,N'-dicarbazolbiphenyl (CBP) doped with 7 wt.-% of the green-emitting phosphor, tris(phenylpyridine)iridium (Ir(ppy)₃). Next, a 4,7-diphenyl-1,10-phenanthroline (Bphen) hole and exciton-blocking buffer layer is deposited, followed by a Li-doped Bphen electron-transport layer (ETL) obtained by co-deposition of pure Li, contributing a free electron concentration of ~10¹⁸ cm⁻³.^[15] The total thickness of the Bphen layers is kept constant at 55 nm, while the thickness of the Li-doped layer varies between 0 and 30 nm. For the device without Li doping, a 0.5 nm thick layer of LiF is deposited prior to the deposition of the Al cathode, which has also been shown to result in n-type conductivity in organic materials such as Bphen.^[16,17]

Conductivity-doped PHOLEDs are fundamentally different from undoped, or adventitiously doped organic electronic devices. One difference is that the electron- and hole-confining

[*] Dr. M. Pfeiffer, Prof. S. R. Forrest,
Department of Electrical Engineering, Princeton University
Princeton, NJ 08544 (USA)
E-mail: forrest@ee.princeton.edu

Dr. M. Pfeiffer, Prof. K. Leo
Institut für Angewandte Photophysik, Technische Universität Dresden
George-Baehr Str. 1, Dresden, D-01062 (Germany)
E-mail: pfeiffer@iapp.de

Prof. M. E. Thompson
Department of Chemistry, University of Southern California
Los Angeles, CA 90089 (USA)

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Table 1. Comparison of the efficiencies of several different phosphor/host combinations reported for PHOLEDs. Here, Φ is the theoretical maximum power efficiency given the emission spectrum of the phosphor, η_p is the measured power efficiency at current density J , $\eta_{Q\text{int}}$ is the internal quantum efficiency, $\eta_{Q\text{ext}}$ is the external efficiency, V_λ is the photon energy divided by the elementary charge at the peak of the emission spectrum, and V is the applied voltage at J .

Phosphor	Host	Φ [lm/W]	$J = 1 \mu\text{A}/\text{cm}^2$				$J = 1 \text{ mA}/\text{cm}^2$			
			η_p [lm/W]	$\eta_{Q\text{ext}}$	$\eta_{Q\text{int}}$	V_λ/V	η_p [lm/W]	$\eta_{Q\text{ext}}$	$\eta_{Q\text{int}}$	V_λ/V
ppy ₂ Ir(acac) [3]	TAZ	53	60	0.19	0.87	0.60	20	0.15	0.68	0.25
btIr(acac) [28,29]	CBP	17	4	0.13	0.65	0.34	2.2	0.10	0.44	0.22
Firpic [30]	CBP	26	1.3	0.06	0.27	0.83	5.0	0.057	0.23	0.34
Ir(ppy) ₃ p-i-n	CBP	53	22	0.05	0.25	1.0	29	0.09	0.45	0.86

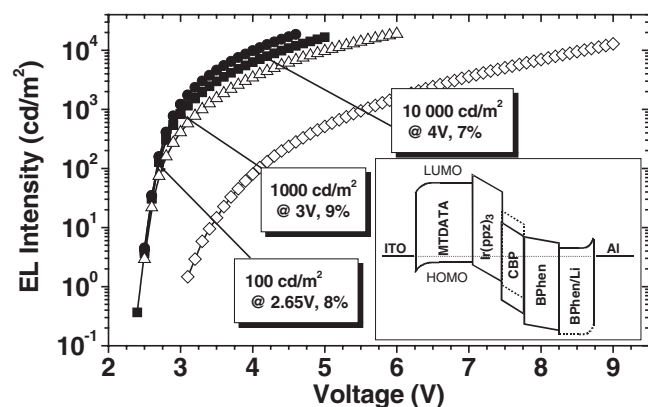


Fig. 1. Electroluminescence (EL) intensity versus applied voltage for CBP:Ir(ppy)₃ based PHOLEDs. Shown are data for p-i-n PHOLED samples A (triangles), B (squares), and C (circles), compared with an undoped, conventional PHOLED (diamonds). The percentages shown in the boxes are the external quantum efficiencies measured at the corresponding voltages. Inset: Proposed equilibrium energy level diagram of the p-i-n PHOLED.

barriers in doped PHOLEDs are due, at least at low current injection, to a combination of built-in potential as well as the energy-level offsets between the adjacent layers. In conventional organic electronic devices, these barriers are solely determined by the energy-level offsets. While at high injection levels this added energy barrier vanishes by application of an external bias, at low levels of injection they can improve charge and carrier confinement, and hence the device efficiency. Furthermore, the very low resistance of the doped layers (typically $< 50 \Omega \text{ cm}^{-2}$) allows for very thick transport layers to be employed. Whereas a typical small-molecule-based device thickness should not exceed 100 nm for the organic material layers, doped devices can be made up to five times thicker without appreciably increasing the voltage drop across the layers. This provides a significant advantage in device yield when large displays are mass produced, due to the elimination of many parasitic shorts between electrodes often encountered in thinner, undoped devices.

The effect of doping the charge-transport layers on the performance of the PHOLEDs is shown in the luminance–voltage (L – V) characteristics in Figure 1. Here, the L – V characteristics of p-i-n PHOLEDs with LiF-doped BPhen (sample A), and a 15 nm and 30 nm thick Li-doped BPhen (samples B and C, respectively) are compared with a conventional, undoped PHOLED structure. The undoped sample

reaches 100 cd m^{-2} at 4 V, with the luminance increase flattening out at higher voltages due to ohmic losses in the transport layers or at injection barriers. Accordingly, a luminance of 10^4 cd m^{-2} is achieved at 8.5 V. In comparison, the intentionally doped p-i-n PHOLEDs operate at a significantly lower voltage, with 100 cd m^{-2} obtained at 2.65 V for samples B and C, increasing to only 4 V at 10^4 cd m^{-2} . A slightly increased voltage is obtained for sample A with a LiF interlayer instead of bulk Li-doping of BPhen, due to the limited diffusion of Li into the BPhen layers from the Al/LiF cathode. The external quantum efficiencies, measured with a calibrated Si detector capturing only light emitted in the forward (viewing) direction, is also shown at several luminances.

While the external quantum efficiencies are typical of conventional CBP:Ir(ppy)₃ PHOLEDs reported previously,^[1] they remain $\sim 50\%$ that of optimized devices, where external efficiencies have been shown to approach $\sim 20\%$.^[3,4] Nevertheless, their luminous power efficiencies exceed that of optimized conventional devices. This improvement, shown in Figure 2, is due to the lower voltage operation of the doped structures. Here, we plot the power efficiency versus current density for sample B, along with those previously reported for optimized, conventional ultra-high efficiency green-emitting PHOLEDs. While the efficiency of the conventional PHOLEDs approaches 70 lm W^{-1} at very low brightnesses, the rapid increase in voltage with current (and hence brightness)

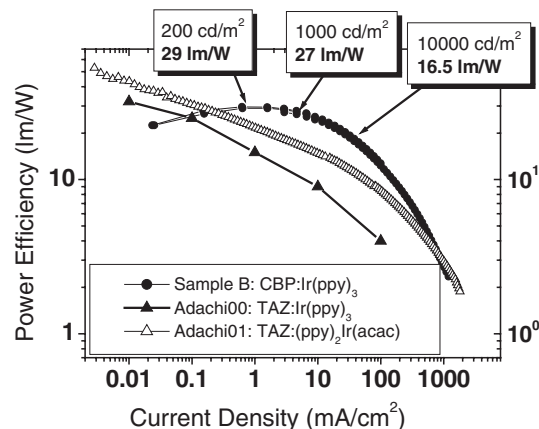


Fig. 2. Luminous power efficiency versus current density for a p-i-n PHOLED sample B, compared to undoped PHOLEDs using a similar CBP:Ir-complex emissive region reported in the literature [2,3].

results in a concomitant rapid decrease in luminance efficiency. Hence, while the power efficiencies of both the doped and conventional PHOLEDs are approximately equal at 100 cd m^{-2} , the doped PHOLED efficiency is much higher as the brightness continues to increase. For example, at 1000 cd m^{-2} , the p-i-n PHOLED has a power efficiency of 27 lm W^{-1} , approximately equal to its efficiency at 100 cd m^{-2} , falling to 16.5 lm W^{-1} at 10^4 cd m^{-2} . This is compared with 29 lm W^{-1} at 100 cd m^{-2} , dropping to 20 lm W^{-1} at 1000 cd m^{-2} , and 13 lm W^{-1} at 10^4 cd m^{-2} for the best conventional, undoped PHOLEDs reported to date.^[3]

Two principle reasons for the low operation voltage in p-i-n-type OLEDs are the efficient injection from contacts into doped transport layers,^[6,9,18,19] and the low ohmic losses in these high conductivity layers.^[20] However, these advantages can only be exploited when using resistive buffer and emission layers with minimal thicknesses. Here, we succeeded in reducing the thickness of the emission layer to only 5 nm without losses in the quantum efficiency. As shown in the inset of Figure 1, a reduction of the EML thickness is made possible using $\text{Ir}(\text{ppz})_3$ as a buffer between the p-doped HTL and the EML. The high LUMO energy of $\text{Ir}(\text{ppz})_3$, combined with the alignment (as determined from its ionization potential) of its highest occupied molecular orbital (HOMO) with that of m-MTDATA,^[12,21] make this material suitable for blocking electron- and exciton-transport, while allowing for efficient hole injection into the EML. Indeed, using $\text{Ir}(\text{ppz})_3$ as an electron blocking layer, we observe only $\text{Ir}(\text{ppy})_3$ electrophosphorescence even with an EML thickness of only 5 nm. Furthermore, the triplet energy of BPhen also lies well above that of $\text{Ir}(\text{ppy})_3$. Combined with its deep HOMO energy of 6.7 eV,^[1] both BPhen and $\text{Ir}(\text{ppz})_3$ provide nearly complete isolation of the EML from the conductive dopants, as well as good confinement of both carriers and excitons in this region, allowing for high efficiencies to be obtained with thin, low resistance EMLs.^[1,2]

The external quantum efficiency as a function of EML thickness is shown in Figure 3. Here, it is clear that the highest efficiencies ($9.5 \pm 0.5\%$) at high current densities are obtained

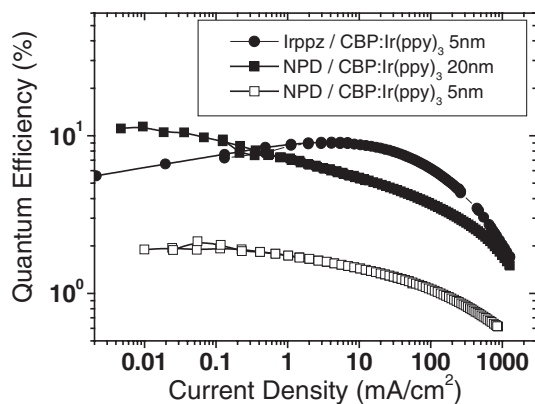


Fig. 3. External quantum efficiency versus current density for the p-i-n $\text{CBP:Ir}(\text{ppy})_3$ PHOLEDs with different emissive-layer thickness (shown in the legend), compared to the efficiency of a thin (5 nm) emissive layer combined with an $\text{Ir}(\text{ppz})_3$ electron and exciton blocking layer.

for an EML thickness of 5 nm. The decrease in efficiency at low currents is attributed to quenching by impurities such as Li atoms having diffused through the buffer layer during the deposition process. In contrast, the efficiencies obtained using the hole-transport material, N,N' -di(naphthalene-2-yl)- N,N' -diphenylbenzidine (α -NPD), in place of $\text{Ir}(\text{ppz})_3$ are considerably lower. This reduction can be reversed to a limited degree if the EML is made thicker (e.g., 20 nm), although this increases the operating voltage of the device, and hence significantly decreases the power efficiency. This is due to the shallow LUMO of α -NPD (approximately 2.6 eV)^[1], which allows generation of excitons on α -NPD from carrier pairs at the α -NPD/EML interface. Moreover, we found that the photoluminescence of $\text{Ir}(\text{ppy})_3$ mixed into α -NPD is strongly quenched due to the lower triplet exciton energy of α -NPD as compared to $\text{Ir}(\text{ppy})_3$. Accordingly, Dexter transfer of triplet excitons from the EML to α -NPD is another possible loss mechanism whenever the exciton generation occurs near to the α -NPD interface.

To understand the conduction mechanism in p-i-n PHOLEDs, we fit the current-density vs. voltage (j - V) characteristics of the doped samples to the Shockley model for a p-n junction:

$$j(V) = j_s \exp \left[\frac{e \left(V - (j/j_T)^{1/m} \right)}{nk_B T} \right] \quad (1)$$

where j_s is the saturation current density, k_B is the Boltzmann constant, T is the absolute temperature, e is the electron charge, and n is the ideality factor. Here, the second term in the exponential accounts for voltage drops across the EML and undoped buffer layers of the device, which are expected to be space-charge limited, where $V_T = (j/j_T)^{1/m}$ is the voltage drop across these layers. For trap-free space-charge-limited currents, $m=2$ and $j_T = 9/8[\epsilon\mu/d^3]$, where ϵ is the permittivity of the layer, d is its thickness, and μ is the charge mobility. Note that $m > 2$ in the presence of traps,^[22,23] or for injection-rather than bulk-limited transport.^[24]

Least-square fits to the forward-biased j - V characteristics for samples A–C in Figure 4 show a reasonable fit to Equation 1 over seven orders of magnitude in current density. The fits yield $n = 2.1 \pm 0.1$, a value typical for inorganic double-heterojunction LEDs.^[25] This is characteristic of recombination of charge carriers within the intrinsic (i) region due to traps, energy wells formed by interfaces, or other recombination mechanisms to be considered in detail elsewhere. Further, we find that $m = 1.7 \pm 0.1$ for these devices, which is close to the value expected for trap-free space-charge-limited transport. The differences in the high current behavior of the three samples is mainly caused by different values for j_T : 10 mA cm^{-2} , 20 mA cm^{-2} , and 42 mA cm^{-2} , for samples A, B, and C, respectively, with the increase corresponding to a decrease in total thicknesses of the undoped region for these samples. The significantly lower value for sample A indicates that free Li from the LiF leads to only a thin doped injection layer while

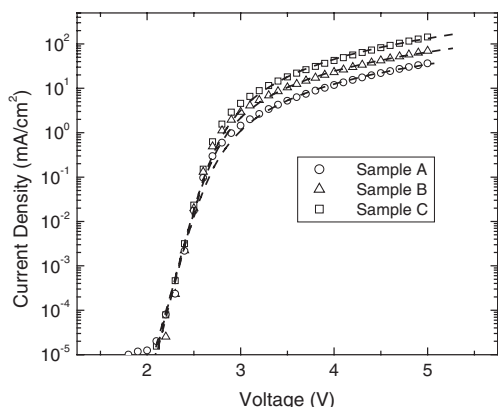


Fig. 4. Current vs. voltage characteristics of three p-i-n CBP:Ir(ppy)₃ PHOLED structures. Also shown are fits to the data assuming a theory combining the Shockley equation [27] and trap-free space-charge limited currents as discussed in text.

the majority of the BPhen remains essentially undoped. To check the fitting parameters obtained from the high current regime, we infer a charge carrier mobility of $\mu \sim 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is consistent with values obtained for most organic amorphous materials. However, this represents a mean value, as the undoped region comprises an electron-transporting, a hole-transporting and a bipolar layer with different mobilities. Using $3 \times 10^{-7} \text{ S cm}^{-1}$ for the conductivity of doped m-MTDA-TA,^[13] the voltage drop over a 50 nm thick layer at $\sim 10 \text{ mA cm}^{-2}$ is $< 0.2 \text{ V}$, and thus is small compared to the total voltage (3 V) applied.

The applicability of the ideal-diode and space-charge-limited model over the device operating range spanning seven orders of magnitude in current, strongly suggests that the limiting mechanisms for carrier transport in doped organic devices are similar to those governing p-i-n junctions employing conventional inorganic semiconductors. That is, carrier diffusion to overcome the built-in energy barrier and subsequent recombination at low voltages, followed by space-charge-limited current due to transport across undoped layers at higher voltages, is similar to the characteristics of a Mott diode. This is in striking contrast to an injection-limited transport characteristic of the undoped layers typically comprising organic thin film devices such as OLEDs.^[24,26]

In conclusion, we have demonstrated very-high-efficiency, conductivity doped p-i-n green-emitting electrophosphorescent OLEDs. By doping the hole- and electron-transport layers, low voltage ($\sim 3 \text{ V}$) operation is achieved, leading to a luminous power efficiency of $> 20 \text{ lm W}^{-1}$ at very high brightnesses. The doped PHOLEDs exhibit nearly ideal diode characteristics up to a current density that generates a brightness of $\sim 1000 \text{ cd m}^{-2}$ at 3 V. At higher brightness, we find that the current is space-charge limited. These results suggest a new architecture for organic electronic devices, where charge confinement is provided by appropriate orbital offsets at heterojunctions, along with the built-in potential due to the Fermi energy offsets of the p- and n-doped regions, and the total undoped region can be kept extremely thin. Nevertheless, doping allows for the use of thick, low resistance layers, with the

potential for increasing yields in large-area organic electronic applications.

Experimental

Here the 50 nm thick (m-MTDA) hole-transport layer (HTL) doped with 2 mol-% 2,3,5,6-tetrafluoro-7,7,8,8-tetracyano-quinodimethane (F₄-TCNQ) is first deposited following procedures described previously [3]. All films are deposited on a pre-cleaned indium tin oxide (ITO) coated glass substrate by vacuum thermal evaporation in a chamber with a base pressure of 10^{-7} torr. Doping is by co-evaporation from independent sources, whose effusion rates are monitored using quartz crystal microbalances. The ITO serves as a transparent anode contact with a sheet resistance of $20 \Omega \text{ sq}^{-1}$.

The materials used in the devices are as follows: m-MTDA = 4,4',4''-tris-(3-methylphenylphenylamino)triphenylamine; F₄-TCNQ = 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane; Ir(ppy)₃ = tris(phenylpyrazole)iridium; CBP = N,N'-dicarbazolbiphenyl; Ir(ppy)₃ = tris(phenylpyridine)iridium; Bphen = 4,7-diphenyl-1,10-phenanthroline; α -NPD = N,N'-di(naphthalene-2-yl)-N,N'-diphenylbenzidine.

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