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High-efficiency green phosphorescence from spin-coated single-layer dendrimer light-emitting diodes

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We demonstrate very high-efficiency green phosphorescence from a single-layer dendrimer organic light-emitting diode formed by spin-coating. A first generation *fac*-tris(2-phenylpyridine) iridium cored dendrimer doped into a wide-gap 4,4′-bis(*N*-carbazole) biphenyl host displays a peak external quantum efficiency of 8.1% (28 Cd/A) at a brightness of 3450 Cd/m^2 and a current density of 13.1 mA/cm^2. A peak power efficiency of 6.9 lm/W was measured at 1475 Cd/m^2 and 5 mA/cm^2. We attribute this exceptionally high quantum efficiency for a single-layer device to the excellent film forming properties and high photoluminescence quantum yield of the dendrimer blend and efficient injection of charge into the emissive layer. These results suggest that dendrimers are an effective method for producing efficient phosphorescent devices by spin-coating. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1469218]

Organic light-emitting diodes (OLEDs) are currently an active area of research due to their applications in flat panel displays. Light-emitting materials for these devices can be divided into two general classes, those that are fluorescent and those that are phosphorescent. Phosphorescent systems are the focus of much recent development, because, in spite of their relatively long-lived emissive state they are able to fully utilize all the charge injected for emission. In contrast in fluorescent systems, triplet formation leads to a substantial loss of efficiency.[1,2] Of the many different phosphor emitters that have been reported for OLED devices, iridium based materials have displayed the most promise due to their high photoluminescence efficiency and relatively short excited state lifetime.[3] Tuning of the emission has been demonstrated by a suitable choice of ligand and emission from the blue through to the red has been described,[3–6] giving the scope for the production of large area full color phosphorescent displays. These highly efficient phosphorescent OLEDs have been formed by evaporation techniques. For green emitting OLEDs, the most efficient devices have been based around *fac*-tris(2-phenylpyridine) iridium [Ir(ppy)$_3$] with power efficiencies of 30–70 lm/W being demonstrated.[3–6] However, these devices have a complicated structure and consist of multiple layers deposited sequentially under high vacuum conditions by thermal evaporation. There is thus scope to simplify the manufacture of these exceptionally efficient devices. Fabrication of OLEDs via solution processible mate-

rials would be simple and also opens up exciting possibilities such as inkjet printing of displays. Previous attempts to solution process phosphorescent devices have incorporated a phosphorescent guest into a polymer host and this has been used with some success in the red[7,8] and green.[9] However, the efficiencies of these devices are still low when compared to evaporated systems.

We demonstrate an alternative approach using dendrimers and show that highly efficient phosphorescence can be obtained from simple single-layer OLED structures. Light-emitting dendrimers consist of a core, dendrons, and surface groups. This modular construction enables processing and electronic properties to be tuned independently. This is a significant advantage over polymeric or molecular materials where attempts to tune the solubility often lead to an undesirable change in the emission properties. The dendritic structure also allows a greater choice of chromophores that in many cases would be unsuitable for isolated use or as a repeat unit of a polymer. There have been reports of both fluorescent[10–13] and phosphorescent[14] dendrimer light-emitting diodes (LEDs), but to date, efficiencies have been relatively low (in the region of 0.1%). In this letter, we present the characteristics of efficient single-layer devices formed by spin-coating new solution processible iridium cored phosphorescent dendrimers. The first (G1) **1** and second generation (G2) **2** dendrimers (Fig. 1) contain a *fac*-tris(2-phenylpyridine) iridium core, phenylene based dendrons and 2-ethylhexyloxy surface groups. In addition, we discuss the effects of dendrimer generation and blends of the

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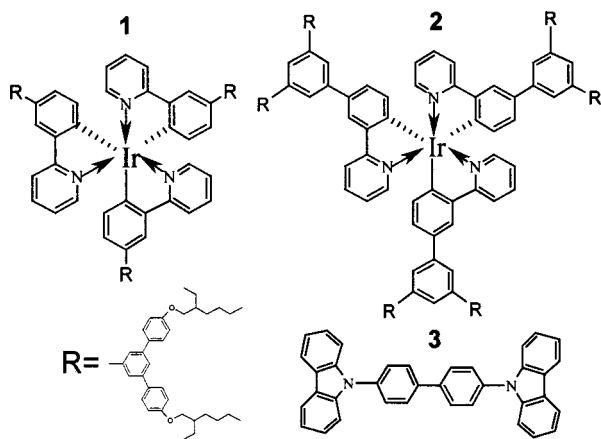[b]Electronic mail: idws@st-andrews.ac.uk

FIG. 1. Structures of the first-generation (G1) **1** and second-generation (G2) **2** iridium dendrimers and CBP **3**.

FIG. 3. External quantum efficiency as a function of current density for the G1, G2, and 20 wt % G1:CBP devices.

material in a host matrix upon the performance of these devices.

Neat films of the two dendrimer generations were compared with a 20 wt % blend of the first generation dendrimer **1** in a 4,4′-bis(*N*-carbazole) biphenyl 3 (CBP) host. The 20 wt % blend of G1 in CBP was chosen because it is equivalent to 6 wt % Ir(ppy)$_3$ which has been shown to be optimal in evaporated LEDs using a CBP host.[3] It was found that these materials could be spin-coated to give good quality films. Films were made using dendrimer solutions of concentration 20 mg/ml in CHCl$_3$ at a spin speed of 2500 rpm for 60 s, giving films with thicknesses of 120–130 nm. In contrast, spincoating a 6 wt % blend of Ir(ppy)$_3$ in CBP was found to produce poor quality films. The absorption and electroluminescence spectra of the two generations are shown in Fig. 2. The absorption spectra can be resolved into absorptions due to the *fac*-tris(2-phenylpyridine) iridium core between 325 and 475 nm and the absorption of the biphenyl dendrons which peak at 272 nm. The latter feature increases with generation due to the larger number of biphenyl units relative to the core present in the G2 material. The electroluminescence (EL) spectrum of G2 peaks at 518 nm and is very similar to that of evaporated devices of Ir(ppy)$_3$ in CBP. The same EL spectrum is observed for the blend of G1 dispersed in a CBP host (data not shown). In contrast the spectrum of the neat
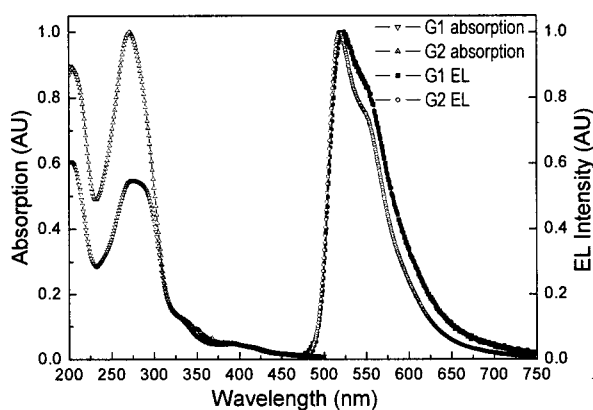

FIG. 2. Absorption (triangles) and EL (squares and circles) spectra of the two generations of iridium dendrimer. Absorption spectra are normalized to the core absorption of the two materials at 400 nm. EL spectra are normalized to 1.
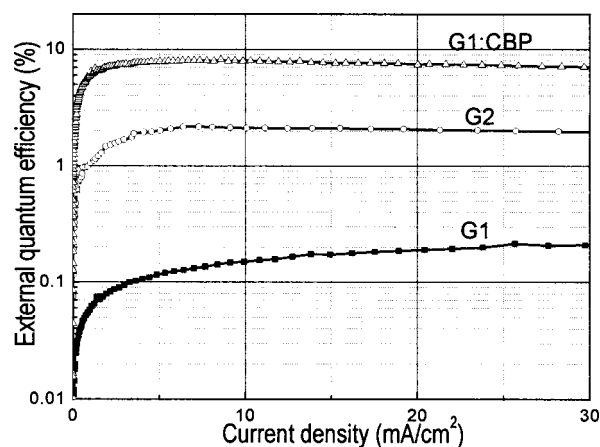
G1 device is slightly redshifted and has an increased red tail. The photoluminescence quantum yields (PLQYs) of the films were measured in an integrating sphere following excitation at 325 nm. For the first generation, the PLQY was found to be 22%. The PLQY was higher at 31% for the second generation and higher still at 78% for the 20 wt % blend of G1 in CBP. The higher quantum yields for the second generation and blend materials can be understood to arise from the greater separation of the Ir(ppy)$_3$ cores, thus reducing concentration quenching effects.

OLEDs were fabricated by spin-coating a 120 nm thick layer of dendrimer onto cleaned and oxygen plasma treated indium tin oxide substrates followed by deposition of 20 nm of calcium capped with 100 nm of aluminum in a vacuum evaporator at a base pressure of $<1\times10^{-6}$ mbar. Current–voltage and brightness–voltage analyses were performed using a Keithley 2400 source measure unit. EL spectra were collected by a charge coupled device spectrometer. The external quantum efficiency (η_{EL}) was calculated by measuring the light output in the forward direction[15] and brightness measurements were also crosschecked with a Minolta LS-100 luminance meter. Figure 3 shows the external quantum efficiencies of three different iridium dendrimer based OLEDs. When the first generation dendrimer 1 was doped into the CBP host at 20 wt %, the OLED had a maximum external quantum efficiency of 8.1% (28 Cd/A) at a brightness of 3450 Cd/m^2. This corresponds to a current density of 13.1 mA/cm^2 and 13.4 V. A maximum power efficiency of 6.9 lm/W at 1475 Cd/m^2 was also observed. This corresponds to a current density of 5 mA/cm^2 and 12 V. The efficiency is lower at low current densities and such an increase in η_{EL} with current density has been observed for evaporated devices with no hole blocking layer[3] and is believed to be due to a saturation of nonradiative sites with increasing exciton density. However, the efficiencies reported for that particular green emitting system were more than one order of magnitude lower than the ones reported here. If the assumption is made that outcoupling losses allow only 20% of the light generated within the film to escape, then we estimate the internal quantum efficiency (η_{int}) to be in the region of 40%. These high efficiencies observed for such a simple device structure are attributed to the excellent film forming properties of the dendrimer preventing recrystallization of
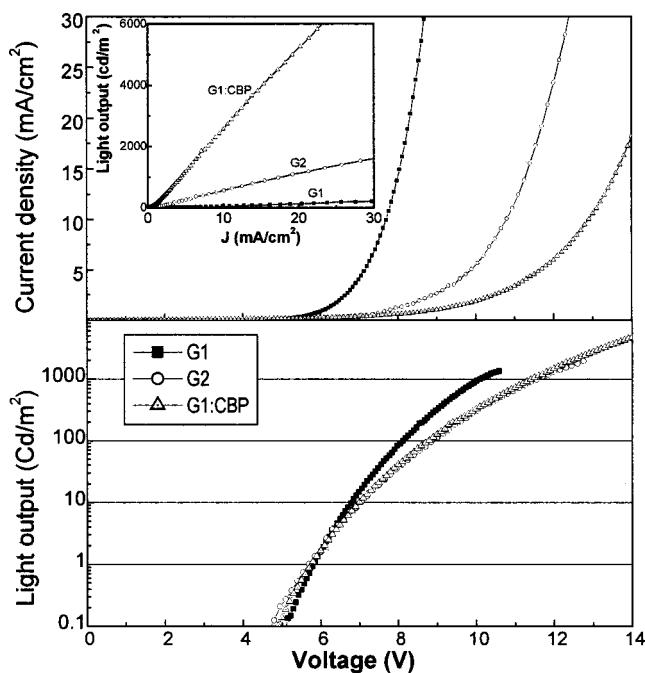
FIG. 4. Current–voltage and light output–voltage characteristics of G1, G2, and 20% G1:CBP devices. Inset: The light output–current density of G1, G2, and 20 wt % G1:CBP devices.

the CBP host, bipolar transport in the film and efficient injection of charge at the anode and cathode. In contrast, a similar device fabricated from 6 wt % Ir(ppy)$_3$ in CBP showed poor stability and device characteristics. A maximum efficiency an order of magnitude lower ($\sim 0.8\%$) was observed and the device was found to be more insulating than that containing the dendrimer blend.

OLEDs based on neat films of the G1 **1** and G2 **2** dendrimers displayed external quantum efficiencies lower than that of the blend device. We found though that there is a striking dependence of the efficiency on generation, with the efficiency increasing by an order of magnitude, from 0.2% to 2.1%, in going from G1 to G2. As the thin-film PLQY only increases by a factor of 1.5 in going from G1 to G2 it is clear that the dendrons have a significant effect on the charge transport within the device. Increasing efficiency with increasing generation has been observed previously in fluorescent dendrimers[10] and attributed to a reduction in the charge mobility with generation. This has the effect of slowing the carriers and thus increasing the probability of exciton formation within the bulk of the device. The light outputs and current–voltage characteristics of all three systems are shown in Fig. 4. The light output–voltage characteristics of the three devices are fairly similar. However, the current for a given brightness is lower in G2 than G1 and lower still for the blend. This accounts for the higher efficiency of G2 than G1, and even higher efficiency of the blend.

To summarize, we have demonstrated that a dendritic molecular architecture can be used to convert molecular compounds which can only be processed by evaporation into materials which can be spincoated to form good quality thin films. We have investigated the effect of generation and shown that the dendritic structure controls core–core interactions and hence the photoluminescence and device properties of the materials. We have also demonstrated high external quantum efficiencies for a simple single-layer LED structure. The results indicate that light-emitting dendrimers are a very promising way of making efficient and easily fabricated spin-coated phosphorescent devices.

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