*Chem. Mater.* **2004,** *16,* 4743−4747

# Ultrahigh Energy Gap Hosts in Deep Blue Organic Electrophosphorescent Devices

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*Received April 12, 2004. Revised Manuscript Received June 24, 2004*

Four ultrahigh energy gap organosilicon compounds [diphenyldi(*o*-tolyl)silane (UGH1), *p*-bis(triphenylsilyl)benzene (UGH2), *m*-bis(triphenylsilyl)benzene (UGH3), and 9,9′-spiro-bisilaanthracene (UGH4)] were employed as host materials in the emissive layer of electrophosphorescent organic light-emitting diodes (OLEDs). The high singlet (∼4.5 eV) and triplet (∼3.5 eV) energies associated with these materials effectively suppress both the electron and energy transfer quenching pathways between the emissive dopant and the host material, leading to deep blue phosphorescent devices with high (∼10%) external quantum efficiencies. Furthermore, by direct charge injection from the adjacent hole and electron transport layers onto the phosphor doped into the UGH matrix, exciton formation occurs directly on the dopant, thereby eliminating exchange energy losses characteristic of guest−host energy transfer. We discuss the material design, and present device data for OLEDs employing UGHs. Among the four host materials, UGH2 and UGH3 have higher quantum efficiencies than UGH1 when used in OLEDs. Rapid device degradation was observed for the UGH4-based device due to electro- and/or photooxidation of the diphenylmethane moiety in UGH4. In addition to showing that UGH materials can be used to fabricate efficient blue OLEDs, we demonstrate that very high device efficiencies can be achieved in structures where the dopant transports both charge and excitons.

## Introduction

Phosphorescent electroluminescent materials and devices are a prime focus of organic light-emitting device (OLED) research due to their ability to efficiently utilize both singlet and triplet excitons.[1] The most common design for phosphorescence-based OLEDs involves a doped emissive region, where the emissive dopant is either an Ir or a Pt complex. The large spin−orbit coupling for these heavy metals leads to efficient intersystem crossing and, hence, short emissive lifetime phosphorescence. To achieve the maximum efficiency in these host−dopant OLEDs, the triplet level of the host must not quench dopant phosphorescence. Such host−guest systems have been realized in green and red organic electrophosphorescent devices, leading to internal quantum efficiencies approaching 100%.[2] Recently, the applicability of host materials with high triplet energies has been demonstrated in blue electrophosphorescent devices, which utilize emission from a bis[2-(4′,6′-difluorophenyl)pyridinato-*N*,$C^{2'}$]iridium(III) picolinate (FIrpic) dopant.[3] Efficient blue electrophosphorescence ($\eta_{\mathrm{ext}} = 7.5\%$) was achieved when using an *N*,*N*′-dicarbazolyl-3,5-benzene (mCP) host, whose triplet level ($E_T = 2.9$ eV) is nearly 0.3 eV above that of FIrpic ($E_T = 2.62$ eV). On the other hand, when an *N*,*N*′-dicarbazolyl-4,4′-biphenyl (CBP) host with a triplet energy of 2.56 eV, ca. 0.06 eV below that of the FIrpic dopant, is used, an efficiency of only 5% is observed. Clearly, the higher triplet energy of mCP is beneficial in achieving the higher efficiency.

To achieve a close match to the National Television Standards Committee recommended blue for video displays, the phosphors used in OLEDs generally need to have triplet energies near 2.8−2.9 eV (emitting at $\lambda \approx 440$ nm). However, as the triplet energy of the phosphor is shifted into the deep blue, it becomes increasingly difficult to find a host with a suitably high energy triplet state. The most common hosts used in phosphorescent OLEDs, carbazole-based materials,[4] have triplet energies of 2.9 eV or less,[5] putting them close to the desired dopant energies. An added complication of deeper blue phosphors is their deep highest

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10.1021/cm049402m CCC: $27.50

occupied molecular orbital (HOMO) levels that typically fall near or below the HOMO levels of the carbazole-based host materials; e.g., mCP and FIr6 (vide infra) have HOMO energies of 5.9 and 6.1 eV, respectively. Having host triplet energies that are close to the phosphor energy and similar host−dopant HOMO levels leads to both energy and electron transfer quenching pathways, which ultimately limit device efficiency.[6]

To circumvent the limitations of carbazole-based hosts, we have studied a series of ultrawide energy gap hosts (UGHs).The materials have large HOMO to lowest unoccupied molecular orbital (LUMO) energy gaps (in the range of 4.5−5.0 eV), and triplet energies greater than that of carbazole (i.e., 3.0 eV). These UGHs are compatible with the organometallic phosphors used in efficient electrophosphorescent devices. It is desirable for the UGH HOMO and LUMO energies to be large enough to lie below and above of the same levels in the dopant, respectively. The guest energy levels are then nested between the HOMO−LUMO gap of the host, thereby eliminating the potential for exciplex formation between the dopant and host. Hence, the guest acts as the primary site for electron and hole conduction within the emissive layer (EML), as well as the trap site for excitons. One example of such a UGH material is polystyrene (PS), which has been used as a host for small-molecule chromophores dispersed by solution processing.[7] However, these PS devices had a single-layer structure, leading to poor charge carrier balance and confinement in the EML, and thus poor efficiency.

In this work, we present a class of small-molecule-based UGH materials applicable to vacuum-deposited OLEDs. We have previously shown that by using ultrahigh energy gap materials it is possible to make highly efficient deep blue phosphorescence devices. By this method, a maximum external quantum efficiency of $\eta_{\text{ext}} = 11.2\%$[8] was achieved using the deep blue phosphorescent iridium complex bis[2-(4′,6′-difluorophenyl)pyridinato-$N,C^{2'}$]tetrakis(1-pyrazolyl)borate (FIr6) as the dopant.[9] Here we discuss the material design, and present device data for OLEDs employing four different UGHs. In addition, we show that high device efficiencies can be achieved in structures where the dopant conducts both holes and electrons in the EML.

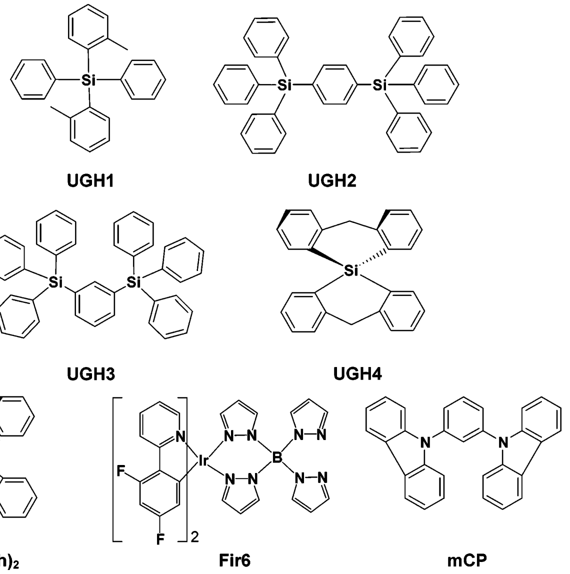

**Figure 1.** Chemical structures of UGH1, UGH2, UGH3, mCP, and FIr6.

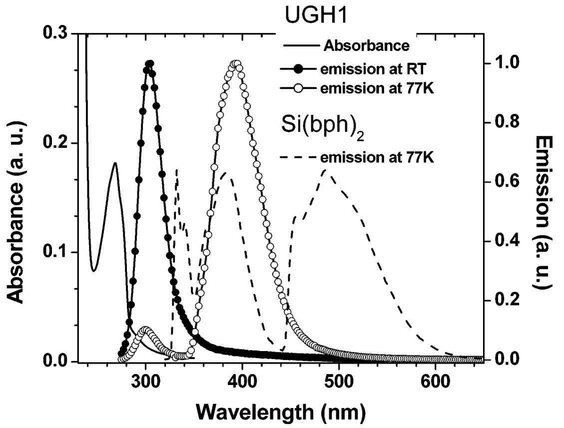

**Figure 2.** Absorption and emission spectra of UGH1 at room temperature and 77 K, and emission spectrum of $Si(bph)_2$ at 77 K.

## Results and Discussion

**UGH Design and Characterization**. Figure 1 lists the chemical structures of diphenyldi(*o*-tolyl)silane (UGH1), *p*-bis(triphenylsilyl)benzene (UGH2), *m*-bis(triphenylsilyl)benzene (UGH3), and 9,9′-spirobisilaanthracene (UGH4). The structures of 5,5′-spirobi(dibenzosilole) ($Si(bph)_2$), mCP, and FIr6 are also shown. Tetraarylsilane compounds have been chosen for two reasons: (i) tetraarylsilicon compounds can be synthesized and modified readily under mild conditions compatible with the demands of organic electronic devices and (ii) tetraarylsilicon compounds can be sublimed without decomposition.[10] The approach taken to maximize the HOMO−LUMO gaps in these materials is to electronically isolate each phenyl ring in the structure, avoiding any conjugating substituents. Direct phenyl−phenyl linkages must be avoided since biphenyl has a triplet energy of 2.8 eV. For example, we have also prepared $Si(bph)_2$, which, due to the biphenyl moieties in the structure, has a triplet energy of only 2.72 eV (Figure 2), making it an unsuitable host for blue electrophosphorescent OLEDs. That is, a blue OLED, having the same architecture as the high-efficiency UGH device,[8] using FIr6 ($E_T = 2.72$ eV, 457 nm) as the dopant and ($Si(bph)_2$) as a host had a maximum external

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(6) An mCP−FIr6-based device is a good illustration of the problems associated with having host triplet energies that are close to the phosphor energy and closely matched host−dopant HOMO levels. Optimized devices with mCP−FIr6 emissive layers give peak efficiencies of only 2.9%, compared to nearly 11% for devices that have both high host triplet energy and a HOMO energy roughly 1 eV below that of FIr6 (see ref 8). The device data for the mCP−FIr6-based devices are given in the Supporting Information.

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quantum efficiency of only $\eta_{ext} = 1\%$. Nevertheless, $Si(bph)_2$ is an efficient host for green phosphorescent OLEDs.[11] Two OLEDs prepared using tris[2-(phenyl)-pyridinato-$N,C^{2'}$]iridium(III) (Irppy; $E_T = 2.46$ eV, 505 nm) as the dopant, one with a doped $Si(bph)_2$ host layer and the other with a doped CBP host layer,[12] gave similar current and luminescence vs voltage characteristics (see the Supporting Information). For example, the Irppy doped, $Si(bph)_2$ host OLED has $\eta_{ext} = 4.5\%$ at a luminance of 10000 cd/m$^2$.

The singlet and triplet energies along with the absolute HOMO−LUMO energies of the UGH materials were determined through a combination of optical and ultraviolet photoemission (UPS) spectroscopies, as described in the Experimental Section. UGH1, UGH2, UGH3, and freshly prepared UGH4 all have nearly identical absorption, room temperature emission, and 77 K photoluminescence (PL) spectra (see Figure 2, showing only the UGH1 spectra as an example). This similarity indicates that little or no conjugation exists between the triarylsilicon groups in UGH2 and UGH3, and between the methylene-linked phenyl rings in UGH4. The singlet energy gaps are estimated to be 4.4 ± 0.1 eV (corresponding to a long-wavelength absorption cutoff of $\lambda = 280$ nm) for each compound, while the triplet energies of the four UGHs are estimated to be approximately 3.5 eV ($\lambda = 360$ nm). Since the redox potentials of the UGH compounds fall outside the range for convenient electrochemical analysis, UPS was used to measure their HOMO energies relative to those in a vacuum. These measurements indicate the HOMO levels of the four UGH materials are at 7.2 ± 0.1 eV.[8] By subtracting the optical energy gap from the HOMO energy, the LUMO levels are estimated at 2.8 eV. The HOMO energies are well below those of other, more conventional materials used in phosphorescent OLEDs, suggesting that UGHs may also block hole injection from the adjacent layer. Furthermore, the LUMO energies of the UGHs fall very near those of the common electron-injecting materials, such as 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) and aluminum 8-hydroxyquinoline ($Alq_3$), allowing for efficient electron injection into the EML (Figure 4).

Two approaches have been adopted to improve the film morphological stability: (i) using Si compounds with multiple arylsilane groups, as in UGH2 and UGH3, or (ii) using nonconjugated linking groups, to prevent ring rotation, as in UGH4. Both approaches lead to an increase in the glass transition temperatures ($T_g$), thus enhancing the likelihood that the materials will form morphologically stable amorphous films.[13] Glass transition temperatures for UGH3 and UGH4 are 46 and 53 °C, respectively, compared to 26 °C for UGH1, whereas UGH2 shows no detectable $T_g$, with a melting point of 345 °C.

The quality of neat thin films of the UGH1 prepared by thermal deposition was investigated by optical microscopy. When UGH1 is deposited directly onto a silicon wafer surface or onto a substrate precoated with an organic film (e.g., *N,N'*-diphenyl-*N,N'*-bis(1-naphthyl)benzidine, NPD), the surface texture appears rough, suggesting the UGH layer has crystallized, as expected for a material with a $T_g$ near room temperature. Codeposition of 92% UGH1 with 8% (by weight) FIr6 inhibits crystallization, such that freshly prepared films are uniform, as expected for an amorphous thin film, although significant surface roughness develops after the film stands for 4−5 h. The surface roughness may be due to the formation of crystalline domains within the doped thin film, which is expected to lead to poor OLED operational stability.[14] Optical microscopy showed that UGH2, UGH3, and UGH4 all give smooth, pinhole-free films, whether they are deposited alone or with a phosphorescent dopant.

**Electrophosphorescent OLEDs Employing UGH Materials**. Blue emitting OLEDs employing the phosphor FIr6 in the EML were fabricated with the structure indium tin oxide (ITO)/NPD (400 Å)/mCP (100 Å)/10% FIr6:UGH (250 Å)/BCP (150 Å)/$Alq_3$ (250 Å)/LiF/Al. Due to the deep HOMO levels of the three UGH materials, it is expected that holes are injected directly onto and carried by the dopant. Consistent with the proposal that the dopant carries charge is the observation that low dopant concentrations in UGH host materials lead to high OLED drive voltages (due to carrier trapping at the dopant), while increasing the doping level gives a marked decrease in drive voltage.[8] Similar dopant-based carrier trapping/transport has been proposed for OLEDs with related wide gap host materials.[15] To increase OLED efficiency, a thin layer of mCP was inserted between the NPD hole transport layer and the EML to reduce the 0.7 eV energy barrier between the HOMO levels of NPD and FIr6, thereby facilitating resonant hole injection[8] into the dopant (see the proposed OLED energy level scheme, Figure 3).[16] In addition, the higher triplet energy of mCP compared to that of NPD ($E_T$ = 2.3 eV[17]) confines excitons within the emissive layer. The external quantum efficiency and current−voltage characteristics of the three UGH devices are shown in Figure 4. All devices have electroluminescence (EL) spectra that match the dopant photoluminescence (PL) spectrum, and at voltages of ~5 V achieve a luminance of 1 cd/m$^2$. The maximum quantum efficiencies of the three devices are 7.0% for UGH1, 9.1% for UGH2, and 8.8% for UGH3. One reason for the relatively low $\eta_{ext}$ of the UGH1 device may be that UGH2 and UGH3 are better solvents for the FIr6 dopant, leading to reduced dopant aggregation, and a concomitantly lower exciton quenching in the EML.

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[11] A recent theoretical study suggested that $Si(bph)_2$ would be a poor host material for blue phosphorescent OLEDs and a good host for green devices. Matsushita, T.; Uchida, M. *J. Photopolym. Sci. Technol.* **2003**, *16*, 315−316.

[12] The device structures used were ITO/NPD (400 Å)/$Irppy_3$:HOST (8%, 250 Å)/BCP (150 Å)/$Alq_3$ (250 Å)/LiF/Al, where NPD = *N,N'*-diphenyl-*N,N'*-bis(1-naphthyl)benzidine, $Irppy_3$ = tris(2-phenylpyridine)iridium, HOST = either $Si(bph)_2$ or CBP, BCP = bathocuproine, and $Alq_3$ = aluminium tris(8-hydroxyquinolate).

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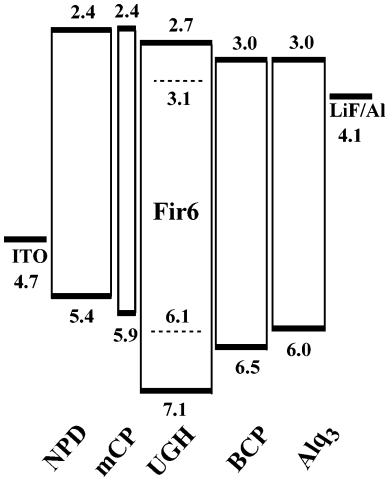

**Figure 3.** Proposed energy level diagram for the following device: ITO/NPD (400 Å)/mCP (100 Å)/FIr6:UGH (10%, 250 Å)/BCP (150 Å)/$Alq_3$ (250 Å)/LiF/Al.

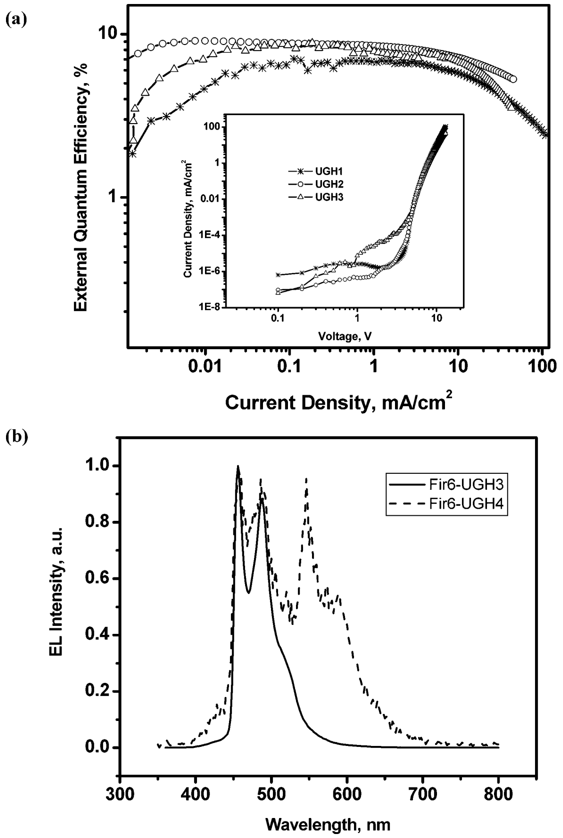

**Figure 4.** (a) External quantum efficiency vs current density for the device NPD (400 Å)/mCP (100 Å)/FIr6:UGH (10%, 250 Å)/BCP (150 Å)/$Alq_3$ (250 Å)/LiF/Al. Inset: current density vs voltage for the same device. (b) EL spectra of FIr6−UGH3- and FIr6−UGH4-based OLEDs, after several minutes of operation. Devices prepared with UGH1 and UGH3 give spectra identical to the one shown for the UGH3-based device. The UGH1, UGH2, and UGH3 devices give the same EL spectrum for extended periods, due to only FIr6 emission. The UGH4-based device initially gives the spectrum shown for the UGH3-based device, but changes to the spectrum shown here occur within a few minutes.

When UGH4 was used as the host, device degradation occurred rapidly during operation and, unlike the behavior of the other UGH devices, an additional broad emission band appeared at $\lambda = 545$ nm at voltages higher than 8 V (see Figure 4b), changing the emission color from blue to yellow-white. The position and shape of the additional peak, together with the fact that device operation was carried out in air, suggest that the rapid spectral changes of the UGH4 device are related to electro- and/or photooxidation of the fluorene moiety, resulting in ketonic defects (i.e., formation of fluorenone groups).[18] This is consistent with the observation that solution PL of UGH4 films exposed to air for a few days also displays a broad peak in the green ($\lambda_{\max} = 515$ nm). We anticipate that the chemical stability of the compound can be significantly improved by replacing the hydrogens in the 9-position of UGH4 with alkyl or aryl groups.

## Conclusions

In summary, we have demonstrated that the use of UGHs can lead to high-efficiency deep blue phosphorescent OLEDs. The ultrahigh energy gaps inherent in these hosts effectively reduce dopant quenching by the host matrix. Furthermore, we demonstrate that ultrahigh energy gap hosts can be "inert", thereby relying on the phosphor dopant to transport both charge and excitons across the EML, resulting in improved blue electrophosphorescent OLED efficiency compared with that of conventional systems relying on host-to-guest energy transfer.

## Experimental Section

UGH1,[19] UGH2,[20] UGH3,[21] UGH4,[22] FIr6,[9] NPD,[23] mCP,[5] and $Alq_3$[24] were prepared according to literature procedures. BCP was purchased from the Aldrich Chemical Co. All materials were purified by temperature gradient vacuum sublimation. Absorption spectra were recorded on an AVIV model 14DS-UV-Vis-IR spectrophotometer and corrected for background due to solvent absorption. Emission spectra (photoluminescence and electroluminescence) were recorded on a PTI QuantaMaster model C-60SE spectrofluorometer, equipped with a 928 PMT detector and corrected for detector response. Solution photoluminescence was taken in 2-methyltetrahydrofuran at both room temperature and 77 K. The HOMO−LUMO gap energy was defined as the point at which the absorption and fluorescence spectra intersect, while the triplet-state energy was determined from the highest energy feature of the low-energy emission band in the 77 K photoluminescence spectra. The HOMO energies for UGH1, UGH2, UGH3, $Alq_3$, mCP, and FIr6 were determined by photoelectron spectroscopy with a He I UV source that has a photon energy of 21.2 eV (Thermo VG Scientific.). The HOMO energies for BCP and NPD were taken from the literature.[25]

Prior to device fabrication, ITO on glass was patterned as 2 mm wide stripes. The substrates were cleaned by sonication in soap solution, followed by boiling for 5 min in trichloro-

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ethylene, acetone, and ethanol in succession for each solvent. Then the substrates were treated with UV ozone for 10 min. Organic layers were deposited sequentially by thermal evaporation from resistively heated tantalum boats at a rate of 2−2.5 Å $s^{-1}$. After organic film deposition, the chamber was vented and a shadow mask with a 2 mm wide stripe was put on the substrate perpendicular to the ITO stripes. Cathodes consisted of a 10 Å thick layer of LiF followed by a 1000 Å thick layer of Al.

The devices were tested in air within 2 h of fabrication. Device current−voltage and light-intensity characteristics were measured using the LabVIEW program by National Instruments with a Keithley 2400 SourceMeter/2000 multimeter coupled to a Newport 1835-C optical meter, equipped with a UV-818 Si photocathode. Only light emitting from the front face of the devices was collected and used in subsequent calculations.

**Acknowledgment.** We thank Dr. David Knowles and Dr. Jie-Yi Tsai for helpful discussions. Financial support was provided by the Universal Display Corp., DARPA, and NSF.

**Supporting Information Available:** Current vs voltage and brightness vs voltage plots for Irppy-doped $Si(bph)_2$ and FIr6-doped mCP-based OLEDs (PDF). This material is available free of charge via the Internet at http://pubs.acs.org.

CM049402M