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We investigate the use of organic surface modifiers based on phosphonic acid anchoring groups that react with the surface of indium tin oxide (ITO) in order to modify its work function, surface energy, and the charge injection in organic multilayer electrophosphorescent devices. The phosphonic acid surface modifiers, possessing different substituting groups, are found to tune the work function of ITO in the range of 4.40–5.40 eV. These surface modifiers have been tested as an interfacial layer between the ITO anode and hole transport layers (HTL) that are either processed from the vapor phase or from solution. The use of this interfacial layer with a solution-processible HTL results in high quantum and luminous efficiencies of 20.6% and 68 cd/A at 100 cd/m^2 (17.5% and 60 cd/A at 1000 cd/m^2). The enhanced performance of the devices incorporating phosphonic acid modifiers could be associated with an improved charge injection and a better compatibility with the hydrophobic nature of the organic layer. The performance of these devices is also compared to that of devices in which ITO is modified with other well-known techniques such as air plasma treatment or the use of a layer of poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate). © 2009 *American Institute of Physics*. [DOI: 10.1063/1.3095492]

I. INTRODUCTION

The interface between electrodes and organic layers in organic light-emitting diodes (OLEDs) has attracted considerable attention because of its importance in controlling the charge injection, thereby impacting the electroluminescence efficiency.[1,2] In order to minimize the energy barrier for charge injection, an anode with high work function and a cathode with low work function are often desirable. Another important factor to consider when compatiblizing the interface between the metal and the organic layer is the surface energy of the electrodes.[3] Indium tin oxide (ITO) is commonly used as the anode due to its commercial availability, good transparency, and low resistivity. Past efforts have sought to modify both the work function and surface energy of ITO by various chemical and physical approaches,[3–5] including conditioning treatments such as air, oxygen, or UV plasma;[6,7] and the use of intrinsically conducting polymers such as poly(3,4-ethylenedioxythiophene)/ poly(styrenesulfonate) (PEDOT:PSS).[8,9] However, each of these approaches presents certain drawbacks. In the case of the aforementioned plasma treatments, the ITO surface becomes more hydrophilic, exhibiting a high surface energy.[4] Additionally, the work function changes obtained through plasma-based treatments are unstable in ambient conditions.[10] PEDOT:PSS is strongly acidic in nature and, when deposited on the surface, can etch the ITO as confirmed by the observed presence of indium in the PEDOT:PSS film.[11] Another important approach for modifying ITO

surface properties is to use monolayers of molecules utilizing various functional groups that can anchor to the surface, such as thiols, silanes, COCl, SO$_2$Cl$_2$, and phosphonic acids.[12–15] Bonding of these molecules to the ITO surface can result in an effective dipole moment at the interface that modifies the work function of ITO, thereby changing the charge injection in a device.[5,13,16] Previous studies have focused on the effects of such interfacial organic surface modifiers on the performance of either single layer diodes or fluorescent OLED devices.[12–15,17–23]

In this paper, we investigate the use of several organic surface modifiers that incorporate a phosphonic acid binding group and their effect on the modification of the work function and surface energy of ITO as well as their influence on the performance of high-efficiency electrophosphorescent OLED devices. The motivation for using phosphonic acid modifiers as compared with modifiers based on other binding groups was their superior bonding ability with hydroxyl-terminated ITO surfaces.[15,16,24] In addition to the effect of the phosphonic acid binding group on the ITO, tuning the organic substituent groups attached to the phosphorus atoms enable one to vary the surface dipole (impacting the ITO work function) and the surface energy. In this study, the performance of these phosphonic acid modifiers as an interfacial layer between the ITO anode and the hole transport layer (HTL) in multilayer electrophosphorescent OLEDs comprised of either evaporated or solution-processible HTLs was tested. A comparison is made with other existing techniques, such as air plasma treatment, and the use of an intrinsically conducting polymer PEDOT:PSS as an injection layer, both of which are often utilized in various organic electronic devices. Modification of the ITO with the phosphonic acid

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a)Author to whom correspondence should be addressed. Electronic mail: kippelen@ece.gatech.edu.
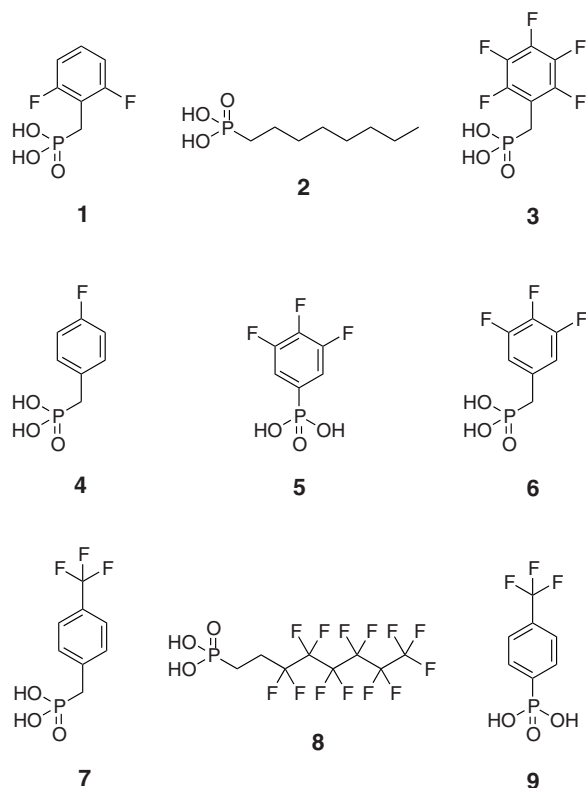
FIG. 1. Chemical structures of the various phosphonic acids used for the surface modification of ITO-coated substrates.
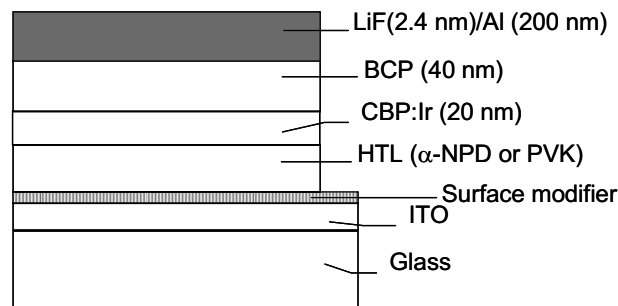

FIG. 2. The device configuration of the electrophosphorescent OLED used to test different surface modifiers at the interface between the ITO anode and the HTL. The HTL was either an evaporated layer of α-NPD (40 nm) or a spin-coated layer of PVK (35 nm).

based modifiers yields high-efficiency devices with both evaporated and solution-processible HTL layers.

II. EXPERIMENTAL

A. Surface modification of ITO

The chemical structures of various phosphonic acids surface modifiers (designated as 1–9) employed in ITO modification are shown in Fig. 1. The phosphonic acids were synthesized following known synthetic routes. Alkyl and benzyl phosphonic acids were synthesized from the analogous starting bromide or iodide. An Arbuzov reaction was performed converting the halide to a diethyl phosphonate. The phosphonate was then converted to the acid using trimethylbromosilane followed by hydrolysis in methanol/water. For the phenylphosphonic acids, the phosphonate was formed from the corresponding halide by either a photoinitiated Arbuzov reaction (9) or a palladium catalyst-assisted reaction (5). Hydrochloric acid was then used to hydrolyze the phosphonate to the acid. All of the phosphonic acids were recrystallized in acetonitrile. Further details and characterization of the compounds will be reported in a separate paper.[25] ITO-coated glass substrates (20 $\Omega/\square$ Colorado Concept Coatings, L.L.C.) were first cleaned in an ultrasonic bath using a dilute solution of Triton-X (Aldrich) in de-ionized (DI) water (20 min) followed by successive ultrasonication in DI water, followed by acetone, and finally in ethanol (20 min each). Washed ITO substrates were then dried in a vacuum oven at 70 °C under vacuum (1×10^{-2} Torr) for 1 h. The surface modification was performed by dipping the cleaned ITO substrates in a solution of phosphonic acid (1 mM in

$CHCl_3:C_2H_5OH::2:1$) for 30 min, followed by annealing of the substrates at 120 °C for 1 h. All the steps were performed in a nitrogen-filled glovebox. The work function values of the modified ITO substrates were measured in air using a Besocke Delta Phi Kelvin Probe apparatus.

B. Device fabrication

To compare the current density-voltage-luminance (J-V-L) characteristics of the OLED devices utilizing different surface modifiers, first a HTL of either evaporated N,N'-diphenyl-N,N'-bis(1-naphthyl)-1,1′ biphenyl-4,4″ diamine (α-NPD) (40 nm) or spin-coated poly(N-vinylcarbazole) (PVK) (35 nm) was deposited onto the modified ITO substrates. An emitting layer of coevaporated (6 wt %) *fac* tris(2-phenylpyridinato-$N,C^{2'}$) iridium in 4,4′-di(carbazol-9-yl)-biphenyl (CBP:Ir(ppy)$_3$) (20 nm) was then deposited at a rate of 1 Å/s. Then, a hole-blocking layer of a bathocuproine [(2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP)] (40 nm) was thermally evaporated at a rate of 0.4 Å/s on top of the emissive layer. All organic materials were thermally evaporated at a pressure below 1 $\times 10^{-7}$ Torr. Finally, an electron injecting layer of 2.4 nm LiF followed by Al cathode (200 nm) using a shadow mask was deposited to yield an active area of $\sim$0.1 cm^2 per device. The final device configuration is depicted in Fig. 2. The devices were tested inside a nitrogen-filled glovebox with O$_2$ and H$_2$O levels <20 and <1 ppm, respectively.

III. RESULTS AND DISCUSSION

A. Influence of surface modifiers on the work function and surface energy of ITO

Figure 3 shows the measured work function values (averaged over three different locations) for ITO substrates modified by various phosphonic acid modifiers, as measured by Kelvin probe experiments. An unmodified ITO substrate exhibits a work function value of 4.50 ± 0.02 eV. It can be seen that a large range of ITO work function values can be achieved through the use of phosphonic acid modifiers containing different substitution patterns (Fig. 1). Octylphosphonic acid (2) does not significantly change the work function in comparison to unmodified ITO. However, pentafluorobenzylphosphonic acid (3), 4-fluorobenzylphosphonic acid (4), 3,4,5-trifluorophenylphosphonic acid (5), 3,4,5-
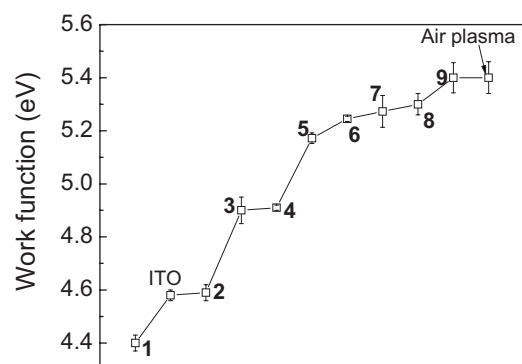
FIG. 3. Work function tunability of the ITO substrate achieved through the use of various phosphonic acid surface modifiers. The work function of unmodified and air plasma-modified ITO is also shown for comparison (the represented values are the average over three different locations on the same substrate).

trifluorobenzylphosphonic acid (6), 4-(trifluoromethyl)benzylphosphonic acid (7), 3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluorooctylphosphonic acid (8), and 4-(trifluoromethyl)phenylphosphonic acid (9), which are partially fluorinated, yield increased work function values with a tunable range of 4.90–5.40 eV (individual values are listed in Table I). The highest achievable work function achieved using a phosphonic acid modifier (9, 5.40 eV) is similar to an air plasma modified work function (5.40 eV). The large increase in the work function due to partially fluorinated modifiers is largely a result of an effective dipole at the ITO surface due to the fluorine atoms. In contrast, 2,6-difluorobenzylphosphonic acid (1) decreases the ITO work function to a value of 4.40 eV, which demonstrates the effect of a dipole pointing in the opposite direction as compared to the other modifiers. Hence, the different substituted patterns of the fluorine atoms in the phosphonic acids can provide a promising tunability in the work function of ITO. This work function tunability can be implemented in a variety of device structures requiring different work functions to match with various organic materials possessing different highest occupied molecular orbital (HOMO) energy levels.[16]

In order to examine the impact of surface modifiers on the hydrophobicity of the ITO substrate, water contact angles were measured (5 μl drop volume). The contact angles (averaged over three different locations) measured on the variously modified ITO substrates are listed in Table I. Unmodified ITO shows an average contact angle of $35 \pm 3°$ and represents the hydrophilic nature of the native surface.[10] Air plasma modification further decreases the contact angle to $<10°$ thus making it more hydrophilic. In contrast, higher contact angles ($>67°$) were measured for all the substrates that were modified with phosphonic acids (Table I). Modifiers 2 and 8 resulted into much higher contact angles, $104 \pm 2°$, and $108 \pm 2°$, respectively. These measurements indicate a fairly hydrophobic surface of the modified ITO, and could provide a better compatibility of the subsequent hydrophobic organic layer deposited on the ITO in a given device geometry.[16]

B. Effect of surface modifiers in evaporated HTL based OLED

The various phosphonic acid surface modifiers were first tested in an electrophosphorescent OLED device geometry comprised of an evaporated α-NPD (HOMO ~5.4 eV) HTL (Fig. 2). The current density (J-V) and luminance-voltage (L-V) measurements of the phosphorescent OLEDs fabricated with differently modified ITO are shown in Figs. 4(a) and 4(b). An unmodified ITO-based OLED showed leakage in the J-V curve which could be associated with the formation of pinholes in the organic layer. The device based on 1, which has a lower work function (4.40 eV), resulted in a similar charge injection as that of unmodified ITO (work function, 4.50 eV), but without any leakage. A similar observation was also seen with 2 as reported elsewhere.[10] The lower leakage J-V characteristics in the case of phosphonic acid modified devices could be due to a better interface between the ITO anode and the organic HTL. The devices utilizing the surface modifiers 3, 5, 6, 8, and 9 also show good diode characteristics with very low leakage. However, the charge injection was higher in the case of OLEDs based on ITO modified with 3, 5, 6, 8, and 9 in comparison to OLEDs based on 1 and unmodified ITO. The J-V characteristics of OLEDs based on ITO modified with 3, 5, 6, 8, and 9 were nearly identical and similar to that of an air plasma treated

TABLE I. Comparison of the work function values measured by Kelvin probe, and the contact angles (values are averaged over three different locations on the same substrate) with water among differently modified ITO substrates. The EQE (%) and luminous efficiency (cd/A) at 1000 cd/m^2 using interfacial surface modifiers in either an evaporated or spin-coated HTL based electrophosphorescent OLED.

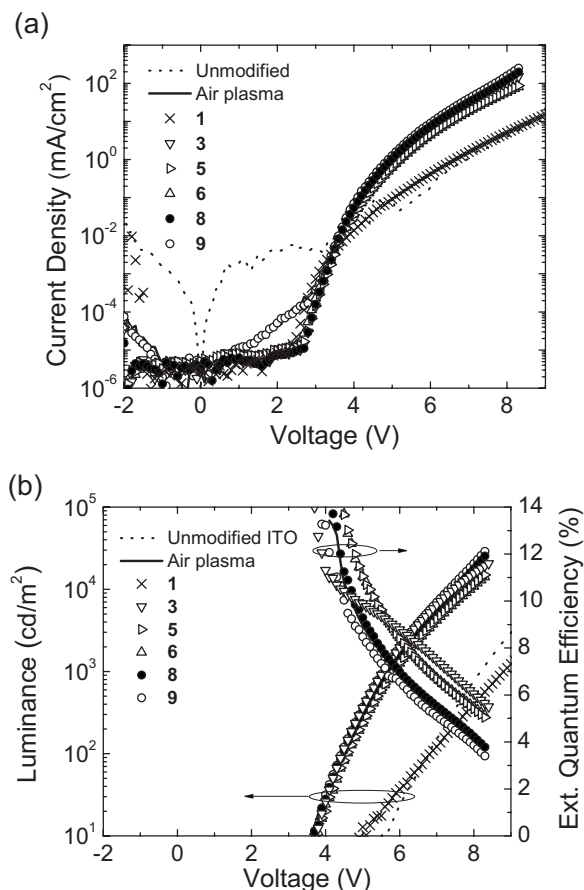| Surface modifier	Work function (eV)	Contact angle	Evaporated α-NPD HTL Efficiencies at 1000 cd/m^2 % (cd/A)	Spin coated PVK HTL efficiencies at 1000 cd/m^2 % (cd/A)
1	4.40 ± 0.03	$75 \pm 4°$	2.4 (8)	$\cdots$
3	4.90 ± 0.05	$70 \pm 6°$	9.3 (32)	17.5 (60)
5	5.17 ± 0.02	$70 \pm 2°$	8.8 (30)	$\cdots$
6	5.20 ± 0.01	$67 \pm 4°$	8.8 (30)	$\cdots$
8	5.30 ± 0.04	$108 \pm 2°$	7.7 (26)	$\cdots$
9	5.40 ± 0.05	$73 \pm 1°$	7.3 (25)	$\cdots$
Air plasma	5.40 ± 0.07	$<10°$	7.6 (26)	17.6 (60)
Air plasma/PEDOT:PSS	5.00 ± 0.04	$\cdots$	$\cdots$	16.9 (58)

(a)

(b)

FIG. 4. (a) Comparison of current density-voltage (J-V) characteristics of ITO/modifier/α-NPD (40 nm)/CBP:Ir (20 nm)/BCP (40 nm)/LiF (2.4 nm)/Al (200 nm) devices with unmodified (dotted line), air plasma modified (continuous line), phosphonic acid modifier 1 (cross symbols), 3 (down triangle), 5 (side triangles), 6 (upward triangles), 8 (closed circles), and 9 (open circles) based OLEDs. (b) The luminance (L) and EQE (%) of the OLED devices with unmodified, air plasma modification, and using various phosphonic acid modifiers [shown in respective symbols as in (a)].

device. Hence, no clear correlation can be seen between the charge injection and the work function among devices with 3, 5, 6, 8, and 9, and the air plasma modification based devices. For example, the device based on ITO modified with 3, which exhibits a work function of 4.90 eV, results in a similar charge injection to that of a device based on ITO modified using 9 (work function=5.40 eV). This could be associated with a pinning effect of the Fermi level of the ITO work function with the HOMO level of the organic layer.[26,27]

The L-V and external quantum efficiency (EQE) curves of the OLEDs as compared in Fig. 4(b) show that the turn-on voltage and the luminance for an OLED incorporating 1 is close to that of an OLED based on unmodified ITO. The EQE curve for the OLED utilizing 1 (not shown for the clarity of the graph) was similar to that of the OLED based on unmodified ITO as reported elsewhere.[10] OLEDs using modifiers 3, 5, 6, 8, and 9 exhibit higher luminance and a lower turn-on voltage (3.7 V) than those using 1 (5.1 V) or unmodified ITO. However, OLEDs based on 3, 5, 6, 8, and 9 themselves exhibited some variation in the EQE curve as a function of applied voltage. OLEDs incorporating ITO modified with 8 and 9 exhibited a lower efficiency than 3, 5, and 6 despite a higher work function of the ITO. EQEs at

1000 cd/m^2 in OLEDs incorporating different phosphonic acid modifiers are listed in Table I. The EQE of an OLED incorporating 8 was 7.7% (26 cd/A) and is comparable to that of one using air plasma treated ITO (7.6%, 26 cd/A). The EQE of an OLED based on ITO modified by 3 was 9.3% (32 cd/A), which is higher than any of the others tested despite 3 imparting only a modest increase in the ITO work function as compared to some of the others. The OLEDs based on modifiers 5 and 6 exhibited an EQE of 8.8% (30 cd/A). Hence, it appears that the EQE and luminance are independent of the work function of ITO when its value is higher than 4.90 e V in the range considered.

C. Effect of surface modification in solution-processed HTL based OLED

In addition to evaporated α-NPD, a spin-coated layer of PVK (HOMO $\sim$5.9 eV) was also tested as HTL layer to investigate the effectiveness of phosphonic acid modified ITO in devices using solution-processed HTLs. Modifier 3 was selected to study these effects because it yielded the best performance in devices based on α-NPD. Here, reference devices incorporating ITO modified with air plasma or with air plasma and a layer of PEDOT:PSS (CLEVIOS PVP AI 4083) were fabricated to compare the effects of surface modifiers with some standard existing ITO surface modification techniques. Figures 5(a) and 5(b) compare the J-V and L-V characteristics of OLEDs incorporating ITO modified with 3, air plasma, and a combination of air plasma/PEDOT:PSS modification procedures. The J-V characteristics of the OLED incorporating 3 show good diode behavior with a high rectification, and a turn-on voltage similar to that of the air plasma-based OLED. Note that OLEDs fabricated with unmodified ITO (not shown here) showed strong leakage and a lower current density than any of the OLED with surface treatment. These results are consistent with the trends observed in the J-V characteristics for devices with α-NPD. The J-V characteristics of the air plasma/PEDOT:PSS modification based OLED were found to be significantly different with a sharp increase in the current at the turn-on voltage and a saturation of the current in a small region before it starts rising again. It can also be noted in Fig. 5(b) that a higher turn-on voltage (6 V) was observed in the PEDOT:PSS modified ITO-based OLED in comparison to that of an OLED using ITO modified with 3 (4.4 V), although the respective work functions of the ITO are quite similar. This increase in the turn-on voltage can be attributed to an increased thickness of the device due to the additional 20 nm thick spin-coated layer of PEDOT:PSS in these devices. Hence, employing a very thin organic modifier such as 3, which does not account for any appreciable change in thickness, at the interface can help keep the turn-on voltage low, which is important in achieving higher luminous efficiency.

It is worth mentioning that air plasma treatments were done just before spin coating of PEDOT:PSS in all devices incorporating this polymer layer. This initial air plasma treatment increases the hydrophilicity of the ITO, improving the wettability of the PEDOT:PSS. When devices were fabricated using PEDOT:PSS modifier without prior air plasma treatment, the devices showed high leakage. On the other
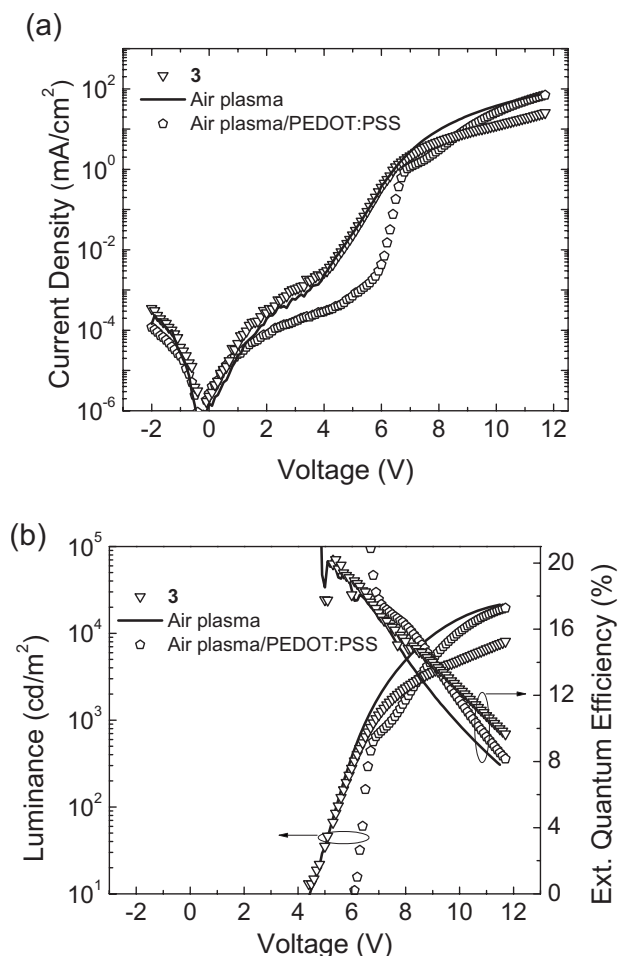
(a)

(b)

FIG. 5. (a). Comparison of the current density-voltage (*J-V*) characteristics of devices with geometry ITO/modifier/PVK (35 nm)/CBP:Ir (20 nm)/BCP (40 nm)/LiF (2.4 nm) /Al (200 nm) OLEDs with modifier 3 (down triangle), air plasma only (line), and air plasma/PEDOT:PSS (pentagon) based modification. (b) The luminance (*L*) and EQE (%) of the OLED devices are shown in respective symbols as in (a).

hand, for organic modifier 3, air plasma treatment is not needed. In addition, there were no added advantages in the work function and *J-V-L* characteristics for the substrates when air plasma treatment was performed prior to ITO modification using 3.

A comparison of luminance and EQE curves of the respective devices as a function of applied voltage is shown in Fig. 5(b) (the values are also summarized in Table I). The EQE at 1000 cd/m^2 for an OLED based on modifier 3 was 17.5% (60 cd/A). At 100 cd/m^2, the OLED with surface modifier 3 exhibited an efficiency of 20.6% (68 cd/A), and is comparable to the OLED incorporating air plasma modified ITO in this study and in previous reports with similar device geometries.[28] These results were highly reproducible across different devices on the same substrate or devices on different substrates fabricated in different runs. In comparison, the EQE of the OLEDs based on air plasma/PEDOT:PSS was 16.9% (58 cd/A) at 1000 cd/m^2 (Table I). Therefore, the modification of the ITO with 3 incorporating a solution-processable HTL results in a device performance comparable to that obtained using the PEDOT:PSS modifier on ITO.

These results are very promising when considering the selection of a surface modification technique in solution-processible OLEDs.

IV. SUMMARY

In summary, phosphonic acid surface modifiers with varying fluorine substitution patterns were used to tune the work function of ITO in the range of 4.40–5.40 eV. These phosphonic acids can be utilized in a variety of device structures that may require tunability of the ITO work function. High efficiencies (20.6%, 68 cd/A at 100 cd/m^2 and 17.5%, 60 cd/A at 1000 cd/m^2) were realized by using ITO modified with phosphonic acids in electrophosphorescent OLEDs based on a solution-processible HTL. This excellent performance of the OLEDs using phosphonic acid modifiers can be attributed to an improved charge injection in comparison to unmodified ITO and a better compatibility with the hydrophobic organic material in comparison to unmodified ITO.

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