

LiF as an n-Dopant in Tris(8-hydroxyquinoline) Aluminum Thin Films**

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Owing to their ease of processing and potential for inexpensive fabrication on low-cost substrates, organic semiconductors have emerged as a novel class of materials for next-generation electronic and optoelectronic devices.^[1,2] The field of organic electronics has generated intense research efforts, spurred on by the promise of a viable alternative to the inorganic materials platform, especially for large-area, flexible electronics applications. Significant progress has been demonstrated towards the realization of efficient organic light-emitting diodes (OLEDs) for flat-panel displays and lighting,^[3] thin-film field-effect transistors (TFTs),^[4] photodetectors for large-area detector arrays,^[5] and organic photovoltaic cells for low-cost solar energy generation.^[5,6] In all of these organic device embodiments, optimization of charge injection/extraction and carrier transport is critically important towards their technological success.^[7] Efficient injection or extraction requires low energetic barriers while competent transport demands highly conductive transport layers.

Organic semiconductors have low carrier concentration and low carrier mobility.^[7,8] Because of these properties, operating voltages of organic devices are generally higher than their inorganic counterparts. Voltage drop in organic transport layers has typically been minimized by using extremely thin structures, which are inevitably more vulnerable to shorts and therefore detrimental to device stability. In addition, because of the low carrier concentration, carrier transport in this class of materials is space charge limited.^[7,8] Similar to inorganic semiconductors, electrical doping can be an approach to enhance the carrier transport, resulting in enhanced device performance. Specifically, doping in organic semiconductors has been employed in organic light-emitting diodes and promised to be an effective way to lower the device operating voltages.^[9] To dope organic semiconductors, typically electron acceptors such as F₄-TCNQ are used as p-dopants^[10] and electron donors such as alkaline metals are used as n-dopants.^[11] While reduced drive voltage and increased brightness have been observed in OLED devices by electrical doping,^[9–11] n-dopants are particularly problematic because of

reactivity of alkaline metals and high diffusivity in an organic matrix.^[9b] In the present study, we demonstrate efficient n-type doping of tris-(8-hydroxyquinoline) aluminum (Alq₃) with inorganic insulator lithium fluoride (LiF) by co-evaporation. We systematically study the effect of dopant concentration on charge carrier transport in this system and show that optimal doping not only leads to enhanced current transport, but also improves electron injection and transport without using low-work-function cathodes. Finally, we employ the optimally doped electron transport layers in OLED architectures to demonstrate devices with enhanced efficiency and lowered operating voltages.

In the first part of the study, single carrier devices were fabricated. Molecular doping was achieved by thermal co-evaporation of LiF and Alq₃. The advantage of such molecular doping is the improved stability compared to alkaline dopants. Insertion of a thin layer of LiF between the organic layer and the metal cathode has previously resulted in reduced driving voltage of an OLED.^[12] In another report,^[13] LiF doping of electron transport layers was shown to improve device efficiency and reliability. Additionally, photoelectron emission measurements have shown^[14] energy-band bending at the Alq₃/LiF interface leading to enhancement in carrier injection at the Alq₃/Al interface. These results indicate a favorable influence of LiF towards improved electron injection and motivated us towards its use as an n-dopant. Typical thin-film device structures consisted of glass substrate/indium tin oxide (ITO)/LiF doped Alq₃/Al. The doping ratio, LiF/Alq₃, was varied from 1 to 15 wt %. In order to compare the transport characteristics of this doping scheme, a device comprising an undoped Alq₃ layer with a LiF/Al electrode, an architecture widely used in OLED devices, was also fabricated. Both device structures are depicted in Figure 1a.

Current–voltage measurements were performed to investigate the effect of LiF doping concentration in Alq₃, if any, on electron injection from the cathode and on the conductivity of the transport layer. Measurements were also done on devices with undoped Alq₃ layer. Figure 1b shows the current density–voltage (*J*–*V*) characteristics of variously doped LiF:Alq₃ devices, along with an undoped device with a LiF/Al cathode. The data clearly show that doping results in devices with significantly higher currents compared to that from the undoped device with bilayer cathode. The inset of Figure 1b depicts a blow-up of the low-current range data for the same devices.

Figure 2a depicts the *J*–*V* characteristics in a log–log plot as a function of doping concentration of LiF in Alq₃. The first point to note is that current density at all operating voltages increases

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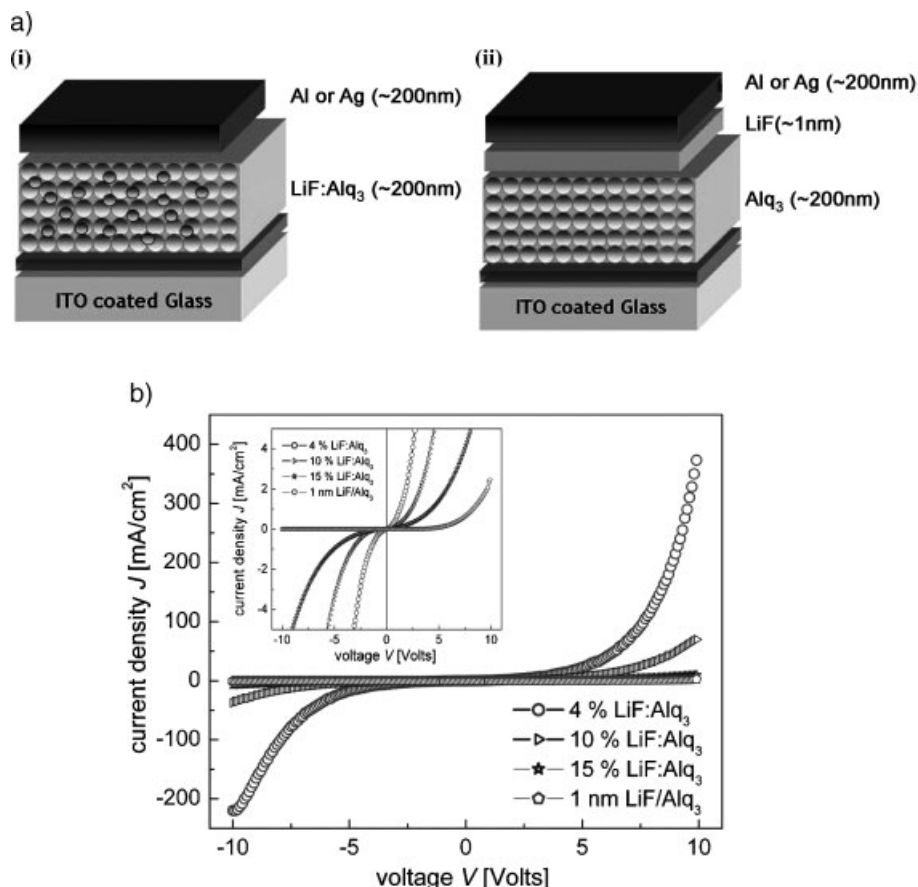


Figure 1. a) Schematic structure of single-carrier devices: i) LiF doped Alq₃ device; ii) Alq₃ device with composite bilayer LiF/Al cathode. b) Current density vs. voltage (*J*–*V*) characteristics for LiF doped Alq₃ devices. Data for a device with LiF/Al bilayer cathode is also shown. Inset: blow-up of the same data in the low-current region to demonstrate the increase in current density with LiF doping.

with LiF doping level, reaching a maximum at a doping concentration of 4–6 wt %. Beyond 10 wt % concentration, the current density decreases with doping concentration, which might be due to aggregation of LiF. At the highest operating bias, the maximum current density in devices with 4% doping is four orders of magnitude higher than that in the undoped device and more than two orders higher than its value in the undoped device with LiF/Al bilayer cathode. The other significant observation is the change in the nature of charge transport with LiF doping. It is clear that in the undoped Alq₃ device and in the one with LiF/Al bilayer cathode, the transport is dominantly trap-limited current (TCL) transport, evident from the $\sim V^{\geq 5}$ dependence^[15] of the current density. The nature of transport changes, interestingly, to a trap-free space-charge limited (SCLC) regime^[15] ($J \sim V^2$) with increasing LiF doping concentration, indicating that traps are being filled by the dopant. Even within the trap-free SCLC regime, a dependence of the current on the doping level is distinctly observed. At voltages below 2 V, the current scales linearly

with applied bias and hence the bulk conductivity (σ) of the thin films can be estimated. Even at a nominal doping of 1%, the room temperature conductivity of the Alq₃ film increases by four orders of magnitude, reaching a value of $5 \times 10^{-7} \text{ S cm}^{-1}$ from $\sim 10^{-11} \text{ S cm}^{-1}$ in the undoped films. At 4% doping concentration, a maximum conductivity of $\sim 10^{-6} \text{ S cm}^{-1}$ is achieved. To estimate the electron mobility in the trap-free SCLC region, we use the Mott–Gurney equation for trap-free SCLC:

$$J = \frac{9}{8} \mu \varepsilon \frac{V^2}{d^3} \quad (1)$$

where ε is the dielectric constant, μ is the carrier mobility, d is the sample thickness. The results show that the low field ($5 \times 10^4 \text{ V cm}^{-1}$) electron mobility in the 4% doped sample is $\sim 1.95 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ which is about two orders of magnitude higher than the value obtained for undoped Alq₃ by time-of-flight (TOF) measurements.^[16] The undoped Alq₃ electron mobility measured by TOF has a strong field dependence and it varies from $10^{-7} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at low fields to $10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at high fields.^[16] Our results indicate that the low field mobility in the doped samples is as high as

the high field mobility of the undoped samples. Table 1 summarizes the conductivities, the carrier concentrations, and the mobility values of both doped and undoped Alq₃ films. Here, the TOF mobility value taken from reference 16 is assumed for the electron mobility of undoped Alq₃ films. The apparent high mobility value estimated by us using trap-free SCLC model in the doped samples indicates that traps are filled, resulting in higher carrier mobility in the doped samples compared to the undoped samples. Additionally,

Table 1. Conductivities, carrier concentrations, and mobilities for both doped and undoped samples.

	Undoped Alq ₃	4% doped Alq ₃
Conductivity [S cm^{-1}]	10^{-11}	10^{-6}
n [cm^{-3}]	4.3×10^{11}	1.5×10^{15}
Mobility [$\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$]	10^{-7} [a]	1.9×10^{-5}

[a] Mobility data taken from Ref. [16c].

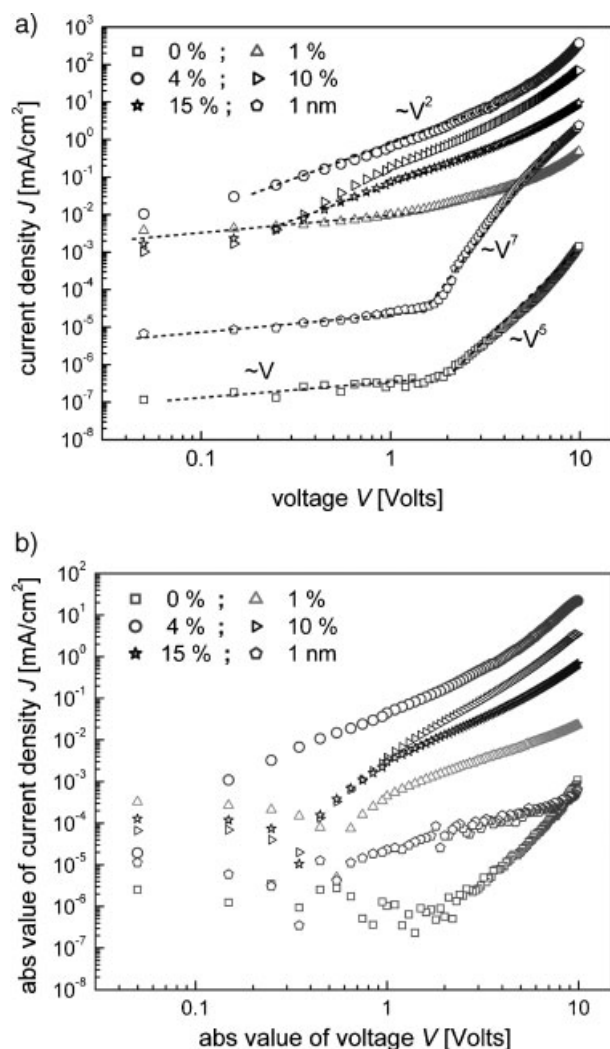


Figure 2. a) Dependence of typical current density–voltage (J – V) characteristics on LiF doping concentration in single-carrier Alq_3 devices with Al cathode. Dotted lines are fits to the data to show the different natures of dependence of current on voltage. b) Similar J – V characteristics parametric in LiF doping concentration for electron injection from ITO. The legends indicate the content of LiF in the samples.

Table 1 also shows that the carrier concentration increases from $4.3 \times 10^{11} \text{ cm}^{-3}$ for the undoped samples to $1.3 \times 10^{15} \text{ cm}^{-3}$ for the doped samples, due to doping.

In order to verify the generality of this approach of n-doping of an electron transporting organic semiconductor to enhance charge injection from cathodes with larger work function, electron-only devices with different metal electrodes were evaluated. Figure 2b shows the current–voltage data from such a device with electron injection from the ITO electrode. The variation of device current with different concentrations of the LiF dopant in Alq_3 is very similar to the case in which Al cathode was employed. Similar trends of increased device current with higher doping level of LiF were also observed in

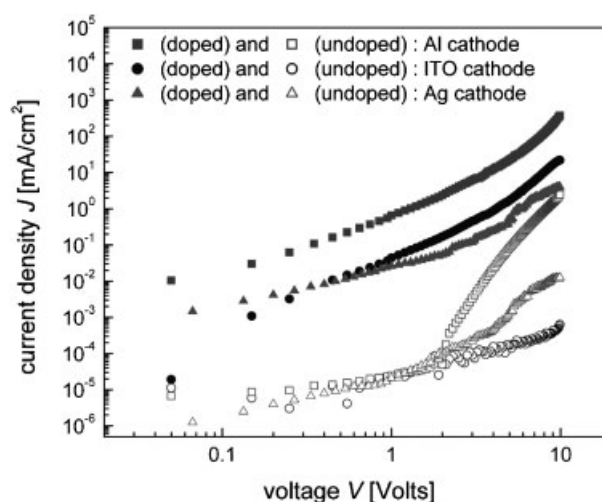
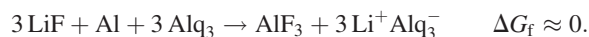


Figure 3. Comparison of efficiency of electron injection into Alq_3 from different electrodes (Al, ITO, Ag) at the optimum concentration of LiF dopant. Data for devices with a bilayer (LiF/electrode) cathode are also shown for comparison. Open symbols correspond to devices with a bilayer cathode, filled symbols refer to doped devices.

another variation of the device employing Ag as the cathode. In Figure 3, the J – V data for electron injection from different types of cathodes into optimally doped devices is compared with corresponding data from an undoped device with a cathode consisting of a thin layer ($\sim 1 \text{ nm}$) of LiF on Al, ITO, or Ag electrode. The results demonstrate that optimally doped devices, with even ITO or Ag electrode, clearly outperform the ones with a thin layer of LiF interposed between undoped Alq_3 and the cathode, in electron injection. This suggests that electron injection^[8,10b] from different metal cathodes is enhanced, as a consequence of the incorporation of LiF into the transport layer. Previous work by Gao and Kahn^[10b] indicated that heavy doping leads to band bending of the organic layer at the metal/organic interface and narrowing of the space charge region (to 2 nm), resulting in more efficient carrier injection. Hence, efficient electron injection from high-work-function electrodes such as ITO and Ag electrode is also possible.

Based on the carrier transport data, it is apparent that LiF doping enhances conductivity in Alq_3 films and it acts as an n-dopant. However, LiF is an insulator and it is not apparent how it acts as an n-dopant in Alq_3 . It has been shown based on the photoemission results from various groups that there is little chemical reaction between LiF and Alq_3 and the possibility that LiF decomposes to form Li^+ to dope Alq_3 can be ruled out.^[17] Then, an obvious question is whether LiF would decompose during the evaporation process. If LiF decomposes during the evaporation process, LiF films deposited by vacuum evaporation should be metallic, or at least show some conductivity. Our data show that evaporated LiF film is a very good insulator with conductivity of $10^{-10} \text{ S cm}^{-1}$ and decomposition of LiF during the evaporation

process is unlikely, which is consistent with the reported photoelectron spectroscopy results.^[17] Previous ultra-violet photoelectron spectroscopy (UPS)^[18] demonstrated that LiF acts as an n-dopant and generates radical anions in Alq₃ in the presence of Al. Specifically, there is direct evidence from conventional^[17a] and synchrotron-radiation^[17b] photoemission spectroscopy results that AlF₃, Li⁺ ion, and n-doped Alq₃ are formed upon deposition of Al on LiF/Alq₃. Results of density functional theory (DFT) calculation also confirm the following reaction:^[17a]



Thus, the formation of radical anion of Alq₃ is responsible for the n-doping effect. Hence, an interfacial layer of LiF^[12] is commonly used to facilitate electron injection from Al into Alq₃. Then the question that arises is whether it is possible that Al atoms are present in the bulk Alq₃ films. Metal diffusion

into small molecule and polymer films^[19] during evaporation has been reported. Based on secondary ion mass spectroscopy (SIMS) measurements, Gradin et al.,^[19a] reported that silver atoms diffuse from the top electrode through the organic layers reaching the bottom ITO anode. In our present experiments, both Al and Ag were used as the top cathode. If Ag can diffuse into the organic layer, it is expected that Al diffusion is even more pronounced given that the evaporation temperature for Al is higher than that for Ag, and diffusion of Al is even more likely as voids or pin holes are formed during the evaporation process. This is probably the reason we experienced a lot of shorting problems in single carrier devices with Al as the top contact metal. Once Al atoms reach the bulk of the Alq₃ layer, LiF molecules might be “activated”^[18] and become active dopants in the Alq₃ film. This model seems to be consistent with our UPS data where we observe only a small shift in the Alq₃ Fermi level when Alq₃ is co-evaporated with LiF. When the Alq₃:LiF film is covered with a sub-monolayer of Al or Ag,

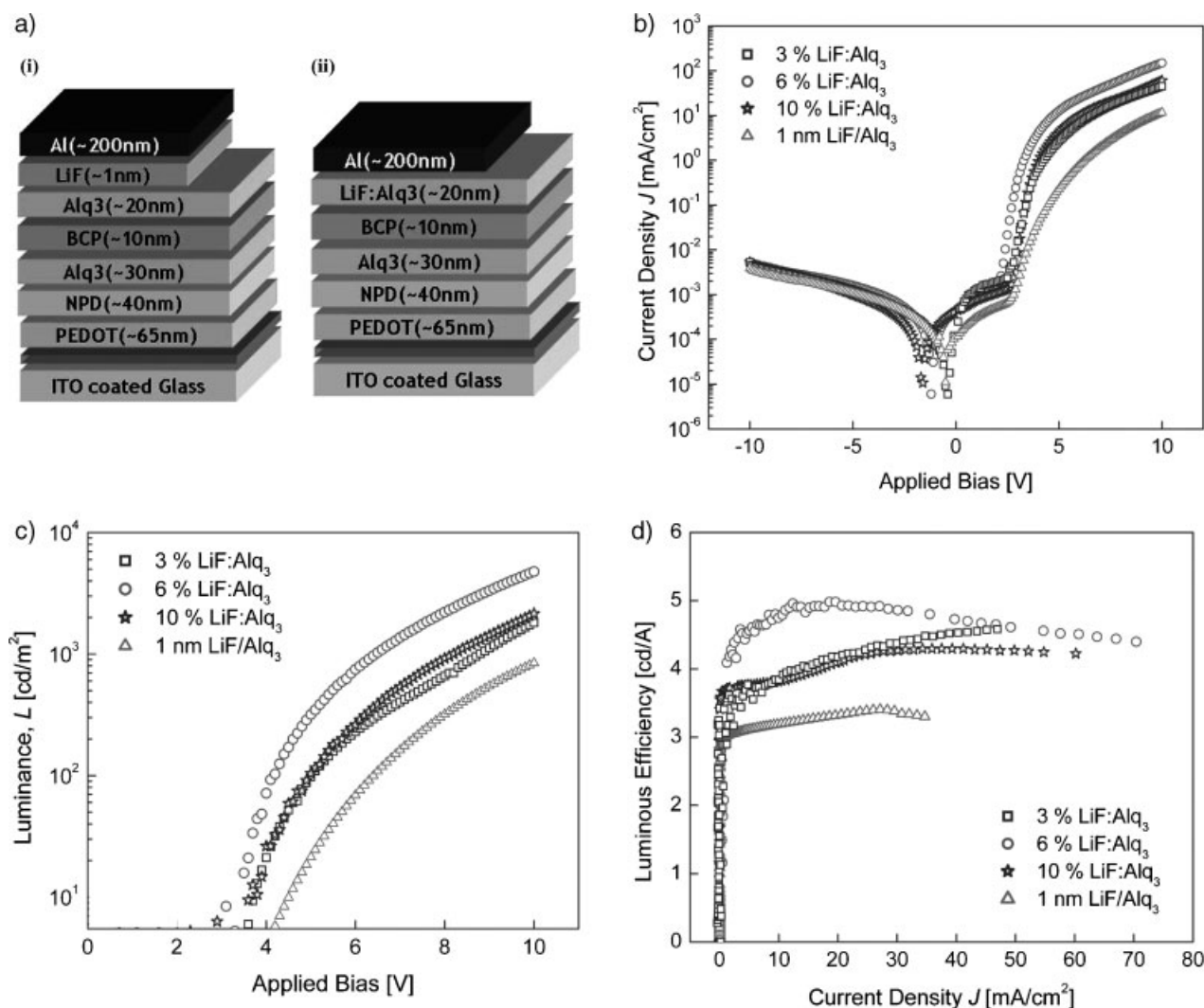


Figure 4. Schematic of OLED structures and device performance as a function of LiF doping in Alq₃. a) Device with (i) LiF doped Alq₃ and with (ii) composite bilayer LiF/Al cathode. b) J - V characteristics. c) Luminance-voltage results. d) Luminous efficiency as a function of driving current density.

a larger shift (1.0 eV for Al and 0.5 eV for Ag) was observed, indicating the doping of LiF in the presence of Al and Ag. Details of the UPS data will be published elsewhere. The fact that we observed doping effect in samples with Ag as the top contact is also consistent with our model since Ag can also activate the doping effect of LiF in Alq₃. Carrier activation by Ag is not as efficient compared to Al, and therefore, the current in the devices with Ag contact is lower than those with Al contact.

Encouraged by the enhanced charge injection and conductivity results, LiF doped Alq₃ electron-injection and transport layer was used to fabricate heterojunction OLEDs. *N,N'*-bis(naphthalen-1-yl)-*N,N'*-bis(phenyl)benzidine (α -NPD) was used as the hole transporting species while another layer of Alq₃ served as the electroluminescent component. A 10 nm thick layer of 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) interposed between the emitting Alq₃ layer and the electron transporting layer serves as an effective hole blocker^[20] to confine the holes in the emitting layer. Typical device architecture is depicted in Figure 4a. The encapsulated devices were characterized for current density–voltage–brightness under ambient conditions. Assuming a Lambertian distribution of emission from the devices,^[21] luminance and efficiency were calculated.

Typical current density–voltage and luminance–voltage characteristics for devices with different LiF doping concentration are plotted in Figure 4b and c, and these are compared to the results from a device with an undoped Alq₃ layer for electron transport with a bilayer LiF/Al cathode. It is obvious from these plots that even low doping levels decrease the operating voltage of the light-emitting devices substantially. All devices with doped Alq₃ layer, for electron-injection and transport, generate significantly larger current density at the same drive voltage compared to the undoped devices. As a consequence, there is marked increase in emission intensity at similar operating voltages. For example, at a driving voltage of 8 V, the luminance of the device with LiF/Al cathode is 325 cd m⁻². For the 3% and 10% LiF doped devices the luminances are 700 cd m⁻² and 950 cd m⁻², respectively, and for the optimally 6% doped device the luminance reaches 2300 cd m⁻² at this applied voltage. Additionally, the devices with doped Alq₃ layer exhibit much lower turn-on voltages. The enhancement of luminance accompanied by a lowering of the operating voltage leads to a maximum increase in the luminous efficiency by almost 70% over a wide range of driving currents in the optimally doped devices as compared to their undoped counterparts; it is demonstrated in Figure 4d. The resulting maximum luminous efficiency of 5 cd A⁻¹ at 19 mA cm⁻² driving current is one of the highest achieved in Alq₃ fluorescent devices with undoped emission layer. In the best devices, this efficiency exhibits only ~10% decrease even at much higher driving currents of ~70 mA cm⁻². The performance of the LiF doped devices is comparable to the alkaline metal-doped devices reported by Kido et al.^[11a] These results clearly demonstrate that doped electron trans-

porting layers, owing to enhanced injection and reduced bulk resistance losses, enable the realization of OLEDs with excellent optoelectronic performance.

In conclusion, we have demonstrated facile non-alkaline n-doping of organic electron transport layers through thermal co-evaporation. Doping of LiF in thin films of Alq₃ increases the current by about six orders of magnitude. Interestingly, doping leads to a gradual filling of intrinsic charge traps in the material, resulting in higher low-field carrier mobility compared to undoped samples. Due to the greatly reduced resistivity, doping leads to enhanced transport and significant decrease in the driving voltage in OLEDs. The use of an inorganic insulator as the dopant instead of reactive alkaline materials traditionally used reduces the concern about reactive dopant diffusion in the organic matrix. The doped transport layer in OLEDs leads to balanced injection and transport of charge carriers, which results in excellent device performance. The technique, if successfully paired with p-type doping of hole-transporting layers, can potentially reduce operating voltage and improve power efficiency of OLEDs even further.

Experimental

Prepatterned ITO-coated glass substrates with a sheet resistance of ~20 Ω per square were cleaned via consecutive treatments in the following sequence: sonication in detergent, de-ionized water, acetone, isopropyl alcohol, rinsing in de-ionized water followed by UV ozone treatment. The organic transporting layer of Alq₃ doped with known proportions of LiF was fabricated by co-evaporation of Alq₃ at 2–5 Å s⁻¹ and LiF at 0.05–0.5 Å s⁻¹ from separate sources at a chamber pressure below 5.0 $\times 10^{-6}$ Torr (1 Torr = 1.333 $\times 10^2$ Pa) to yield a 200 nm thick active layer. The doping ratio, LiF/Alq₃, was varied from 1 to 15 wt %. Typical single-carrier device structures consisted of glass substrate/ITO/LiF doped Alq₃/metal cathode (Al, Ag). Another set of devices comprising undoped Alq₃ and a bilayer LiF/Al or LiF/Ag cathode was also fabricated. Current–voltage (*I*–*V*) measurements were carried out on these devices at ambient temperatures using a Keithley 2400 source meter.

For the light emitting devices, all organic materials were used for device fabrication as purchased without any further purification. Typically, a 65 nm layer of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS; Baytron PVP 8000) was spin-coated onto the cleaned ITO substrate and annealed at 180°C for 15 min in ambient. This serves to planarize the ITO surface, eliminate unwarranted shorts, and facilitate better hole injection. The rest of the small-molecule organic layers in the OLED were vacuum-deposited sequentially at a base pressure of 2 $\times 10^{-6}$ Torr to yield the desired device architecture. Devices were encapsulated under N₂ using cover glass lids and UV-curable epoxy. Current–voltage–emission of the devices were measured on a test fixture using Keithley 2400 source meter interfaced with Labview for data acquisition and a Si photodiode calibrated for the relevant spectral range. The turn-on voltage was defined as the applied bias when emitted light is first detected. This clearly resolved detection limit of emission was set to 8 cd m⁻².

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