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Efficient polymer light-emitting diode with air-stable aluminum cathode

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The fast degradation of polymer light-emitting diodes (PLEDs) in ambient conditions is primarily due to the oxidation of highly reactive metals, such as barium or calcium, which are used as cathode materials. Here, we report the fabrication of PLEDs using an air-stable partially oxidized aluminum (AlO_x) cathode. Usually, the high work function of aluminum (4.2 eV) imposes a high barrier for injecting electrons into the lowest unoccupied molecular orbital (LUMO) of the emissive polymer (2.9 eV below the vacuum level). By partially oxidizing aluminum, its work function is decreased, but not sufficiently low for efficient electron injection. Efficient injection is obtained by inserting an electron transport layer of poly[(9,9-di-n-octylfluorenyl-2,7-diyl)-alt-(benzo[2,1,3]thiadiazol-4,8-diyl)] (F8BT), which has its LUMO at 3.3 eV below vacuum, between the AlO_x cathode and the emissive polymer. The intermediate F8BT layer not only serves as a hole-blocking layer but also provides an energetic staircase for electron injection from AlO_x into the emissive layer. PLEDs with an AlO_x cathode and F8BT interlayer exhibit a doubling of the efficiency as compared to conventional Ba/Al PLEDs, and still operate even after being kept in ambient atmosphere for one month without encapsulation.

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I. INTRODUCTION

Since the discovery of polymer light-emitting diodes (PLEDs), extensive studies have been carried out to optimize their performance.¹ Next to performance, a major issue of PLEDs is their limited stability in ambient conditions. When freshly fabricated PLEDs are placed in normal ambient, black spots start to grow immediately due to the reaction of chemically reactive electrode materials, such as Ca or Ba, with oxygen or water vapor.²

Since the lowest unoccupied molecular orbital (LUMO) of organic electroluminescent materials mostly lies between -2.0 eV and -3 eV, low work function electrodes like Ba (2.7 eV) or Ca (2.9 eV) are typically used as electron injection electrodes, such that electron injection is not hindered by energy barriers. However, metals with such low work functions are very reactive, and the presence of water vapor and oxygen causes device degradation almost immediately when the un-encapsulated device is exposed to air. As alternatives for Ba or Ca, also other electrode materials based on salts as cesium stearate³ or lithium fluoride (LiF)⁴ have been used. Furthermore, to enhance electron injection from more stable electrodes with a higher work function, as aluminum (Al), organic electron transport layers as tris-8-hydroxyquinoline aluminum (Alq_3) and conjugated polyelectrolytes were inserted in between the cathode and the emissive layer.⁵⁻⁹ Although some of these alternatives were able to reach a comparable electron injection efficiency as Ba or Ca cathodes, the PLED lifetime in ambient was not reported. To make the PLEDs intrinsically more robust against water and

oxygen, meaning that in a production line there is more flexibility with regard to PLED fabrication and subsequent encapsulation, other methods were employed to delay the ingress of O_2 and H_2O towards the reactive cathode by using very dense Al capping layers, in order to retard dark spot growth with methods like ion beam-assisted deposition.¹⁰ However, this is only a temporary remedy, since reactive metals are still present in the PLED device structure.

In this study, we introduce partially oxidized aluminum (AlO_x) as an alternative for reactive metals like Ca or Ba in the top electrode. In order to obtain efficient electron injection, an additional electron-transport layer of poly[(8,9-di-n-octylfluorenyl-2,7-diyl)-alt-(benzo[2,1,3]thiadiazol-4,8-diyl)] (F8BT) is inserted between the AlO_x cathode and the emissive polymer. Next to forming an energetic staircase for electron injection, the hole-blocking effect of the F8BT layer also reduces quenching of excitons at the cathode. As a result, the efficiency of PLEDs with an air-stable AlO_x cathode is two times larger as compared to conventional single-layer PLEDs with a Ba/Al or Ca/Al cathode.

II. EXPERIMENT

To study the transport of holes and electrons individually, single-carrier devices were fabricated. Glass substrates and glass substrates pre-patterned with indium tin oxide (ITO) were cleaned using a standard procedure including scrubbing, washing, sonicating in acetone and isopropanol, drying, and UV-ozone treatment. To fabricate electron-only devices, the polymer was sandwiched between an aluminum bottom electrode and an aluminum (or partially oxidized aluminum, AlO_x) top electrode on a glass substrate. To fabricate an Al cathode, aluminum is typically evaporated at partial pressures of O_2 and

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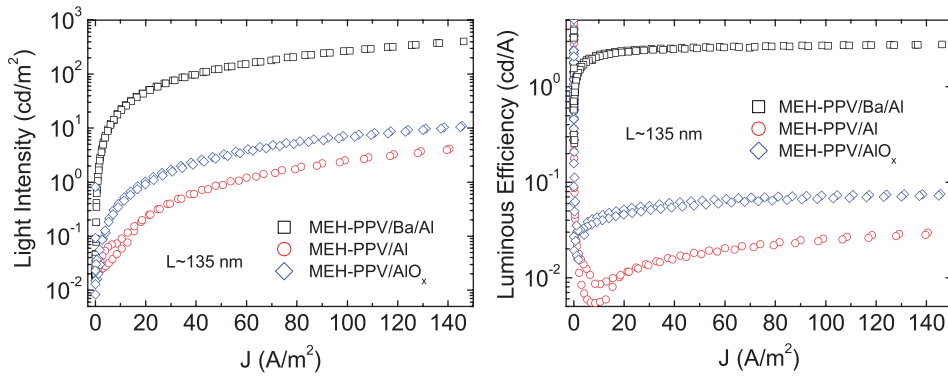


FIG. 1. (a) Light-output versus device current and (b) luminous efficiency of the single-layer and with Ba/Al, Al, and AlO_x cathodes.

H_2O of 1.5×10^{-8} Torr and 0.5×10^{-9} Torr, respectively. To achieve a partially oxidized aluminum (AlO_x) thin film directly on top of the organic, we start evaporation of Al when the partial pressures of H_2O and O_2 in the evaporation chamber are about 1.5×10^{-7} and 1.5×10^{-8} Torr, respectively. For measuring hole transport, hole-only devices were fabricated by sandwiching the polymer layer(s) between PEDOT:PSS (Clevios™ P VP Al 4083) and MoO_3/Al electrodes on top of ITO patterned glass substrates. PLEDs or double-carrier devices were made by sandwiching polymer layers between ITO/PEDOT:PSS as anode and Ba/Al or partially oxidized aluminum (AlO_x) as a cathode. Single-layer PLEDs were spin-casted from dissolved poly[2-methoxy-5-(20-ethylhexyloxy)-p-phenylene vinylene (MEH-PPV) in chlorobenzene as the emissive layer. For bilayer PLEDs, F8BT is spin-casted on top of MEH-PPV layer from toluene solution as the electron transporting layer (ETL).

III. RESULTS AND DISCUSSION

MEH-PPV is a model polymer that has been extensively investigated in the last two decades. Its physical parameters are well known, and therefore, we use MEH-PPV as emissive layer and the green emitting F8BT as hole blocking layer, because its HOMO (-6 eV) is much deeper than MEH-PPV (-5.2 eV).

The first PLED reported in 1990 consisted of a PPV thin film that was sandwiched between ITO and Al cathode. Due to the inefficient electron injection from Al the PLED had a low efficiency.¹¹ Later on, by using PEDOT:PSS as hole injection layer and Ba(or Ca)/Al as cathode, the efficiency of PLEDs based on PPV derivatives increased by almost 1000 times. However, a disadvantage of using Ba or Ca as cathode is that they are chemically very reactive, leading to fast degradation of the PLED after fabrication. A cathode with an equivalent injection electron efficiency combined with air stability would strongly improve the stability and flexibility during fabrication. Here, we use partially oxidized aluminum (AlO_x) as cathode, which is air-stable. It should be noted that the work function of fully oxidized aluminum (Al_2O_3) is 1.3 eV,¹² while the work function of pristine aluminum amounts to 4.2 eV. Now with partial oxidation, the work function of aluminum can be adjusted in the range of 3.5 eV–3.7 eV, which can be used as cathode for electron injection in PLEDs. The work function of AlO_x was verified by Kelvin probe measurements.

As a first step, we compare the light output and luminous efficiency for MEH-PPV diodes with three different cathodes, consisting of Ba/Al, Al, and AlO_x , respectively, as shown in Figs. 1(a) and 1(b). The PLED with Ba/Al shows the highest light output and efficiency (2.7 cd/A), whereas the PLED with an Al cathode shows the lowest values for light output and efficiency (0.026 cd/A). This very low luminous efficiency is a result of the presence of an electron injection barrier due to the high work function offset between Al and MEH-PPV. For the PLED with an oxidized Al cathode, the light intensity and device efficiency (0.072 cd/A) are slightly improved. To improve the device performance further, we have used an intermediate electron transport layer with its LUMO between the Fermi level of the oxidized aluminum cathode and the MEH-PPV LUMO. For this reason, we selected the polymer F8BT, having its LUMO at -3.3 eV, which is situated halfway between the MEH-PPV and AlO_x levels. Furthermore, F8BT has a deep HOMO (-6 eV), such that it efficiently blocks holes that are injected from the anode into the MEH-PPV emitting layer, as schematically shown in Fig. 2.

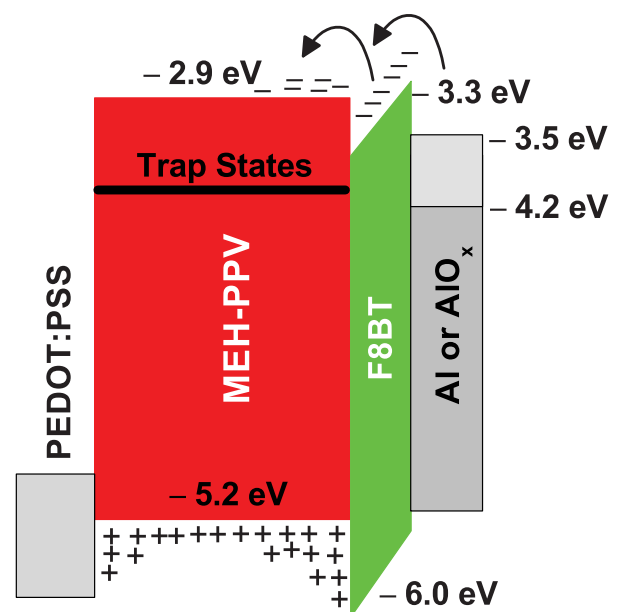


FIG. 2. This sketch shows the energy diagram for fabricated bilayer devices in which the LUMO of F8BT forms an energetic staircase for electron injection from cathodes like Al (-4.2 eV) or AlO_x (~ -3.5 eV). Ba/Al can form an Ohmic contact with either MEH-PPV or F8BT.

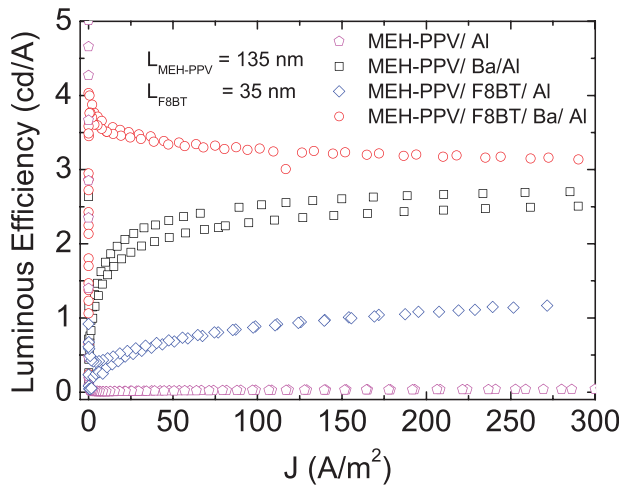


FIG. 3. Luminous efficiency of single-layer MEH-PPV and bilayer MEH-PPV/F8BT devices with both Al and Ba/Al cathodes.

To observe the effect of an F8BT hole-blocking layer on electron injection and consequently the PLED efficiency, we fabricated MEH-PPV single-layer PLEDs and MEH-PPV/F8BT bilayer PLEDs, with pristine Al and conventional Ba/Al cathodes. The device efficiencies are shown in Fig. 3. As mentioned previously, the single-layer PLED with Al cathode is relatively inefficient, reaching 0.026 cd/A at 100 A/m². Insertion of a Ba interlayer (5 nm) enhances the single-layer PLED efficiency to 2.5 cd/A due to the enhanced electron injection. We also observe that with the same cathodes, the double layer MEH-PPV/F8BT PLEDs exhibit a higher efficiency as compared to their single-layer MEH-PPV counterparts. A reason for this efficiency enhancement is that due to the insertion of a hole-blocking layer the recombination zone is moved away from the metallic cathode, thereby reducing the quenching of excitons via energy transfer.^{13,14} Furthermore, the efficiency enhancement is much larger for the Al cathode, from 0.026 cd/A for the MEH-PPV/Al PLED to 0.9 cd/A for the MEH-PPV/F8BT/Al double layer PLED. This strong improvement in efficiency results from a better electron injection in the bilayer device due to the presence of an energetic staircase. In addition, due to hole-blocking at the MEH-PPV/F8BT interface, an electric field across the F8BT layer builds

up, which also improves the electron injection. As a next step, we use the combination of a bilayer structure with a partially oxidized AlO_x cathode.

To separately test the electron and hole injection, we studied the hole and electron current in single-carrier devices, both in the single- and bilayer structure. Fig. 4(a) shows that the hole current in the MEH-PPV/F8BT bilayer device— injection from the PEDOT:PSS anode—is almost three orders of magnitude lower as compared to the MEH-PPV single-layer device. This shows that F8BT, indeed, efficiently blocks holes at the MEH-PPV/F8BT interface due to the offset in HOMO energy. Fig. 4(b) shows the electron current measured for single-layer and bilayer devices with Ba/Al and AlO_x cathode. For the Ohmic Ba/Al contact, we observe that upon addition of 35 nm F8BT to the 135 nm MEH-PPV layer, the current drops by a factor of 10. This drop is mainly due to the increase of the total device thickness. The trap-limited electron current has a very strong thickness dependence,¹⁵ typically given by $L^{-(2r+1)}$. Here, the value of r can be directly determined from the slope of $\log J$ - $\log V$ at higher voltages. Typically, for MEH-PPV $r=4-5$, whereas for F8BT $r=3$ has been reported.¹⁶ Taking $r=4$ ($2r+1=9$) is expected to lead to a reduction of the current of $(135/170)^9$, which is equal to 8. This is very close to the experimentally observed current drop of a factor of 10. An additional contribution for the decrease in electron current might originate from the small LUMO offset between MEH-PPV and F8BT. Electron-only devices with pure aluminum as the injecting cathode show very low currents and are strongly injection-limited (not shown here). Most importantly, the bilayer PLED with AlO_x cathode shows an electron current that is already close to the current of the bilayer device with Ba/Al cathode, especially at higher voltages. For a bilayer PLED, the electron injection from the AlO_x cathode can be even further improved due to accumulation of holes at the MEH-PPV/F8BT interface, creating an increased electric field that enhances electron injection.

To test the performance of AlO_x in PLEDs, the current, light-output, and luminous efficiencies of a reference single-layer MEH-PPV device with Ba/Al cathode were compared with bilayer PLEDs with Ba/Al and AlO_x cathodes. As

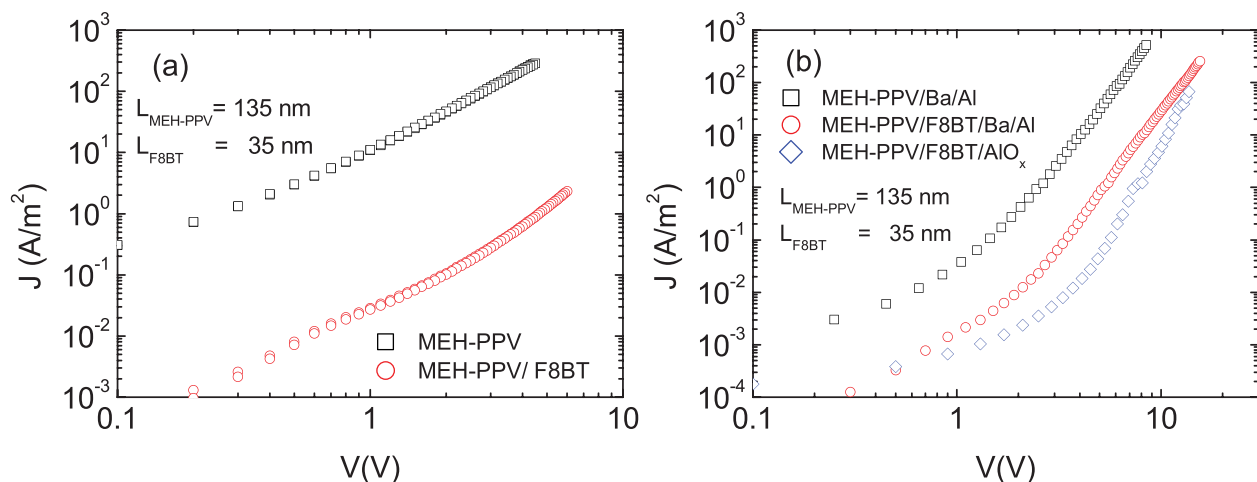


FIG. 4. (a) Hole transport in single-layer and bilayer PLEDs, (b) electron transport in electron-only devices with different cathodes.

shown in Fig. 5, the best results were observed for bilayer devices, exhibiting an efficiency increased up to a factor of 2 in comparison with the reference single-layer PLED with Ba/Al cathode. Surprisingly, the device with an AlO_x cathode is even more efficient than the bilayer PLED with a Ba/Al cathode. To the best of our knowledge, this is the first time that a PLED with an air-stable cathode outperforms a similar PLED with a reactive Ba/Al cathode. Figs. 5(a) and 5(b) show a drop in current and light intensity, which, as already mentioned, is mainly related to the PLED thickness increase upon addition of the F8BT layer. In Figs. 5(c) and 5(d), it can be observed that both the bilayer devices with Ba/Al and AlO_x cathode have a higher efficiency as compared to the MEH-PPV/Ba/Al reference device. This enhanced efficiency is due to reduced quenching of excitons at the cathode, as well as a reduction of the trap-assisted recombination in the emissive layer, as explained in the following. The emissive Langevin recombination R_L is proportional to the sum of electron and hole mobility multiplied by electron and hole density in the recombination zone,¹⁷ whereas nonradiative trap-assisted (Shockley-Read-Hall) recombination R_{SRH} is dependent on electron trap density, hole mobility and hole density as given below

$$R_L = \frac{q}{\varepsilon_r \varepsilon_0} (\mu_n + \mu_p) (np - n_i^2), \quad (1)$$

$$R_{SRH} \approx \frac{q}{\varepsilon_r \varepsilon_0} \mu_p N_t p, \quad (2)$$

where q is the elementary charge, ε_r and ε_0 are the relative and vacuum permittivity of the polymer, μ_n and μ_p are the electron and hole mobility, n and p are the free electron and hole density, n_i is the intrinsic carrier concentration, and N_t is the electron trap density.

Due to trap-limited electron current in a single-layer PLED, the recombination zone will be limited to the region near the cathode. In this region, far away from the hole-injecting anode, the hole density is relatively low. However, upon addition of a blocking layer, the density of holes strongly increases in the emissive region, which is now at the MEH-PPV/F8BT interface. The accumulation of holes also enhances the amount of electrons at this interface due to neutralization effects. In fact, the density of electrons and holes becomes more or less equal at the polymer/polymer interface, as also shown by Coehoorn *et al.*¹⁸ Thus, at the MEH-PPV/F8BT interface the densities of both holes and electrons are enhanced, leading to a stronger enhancement in

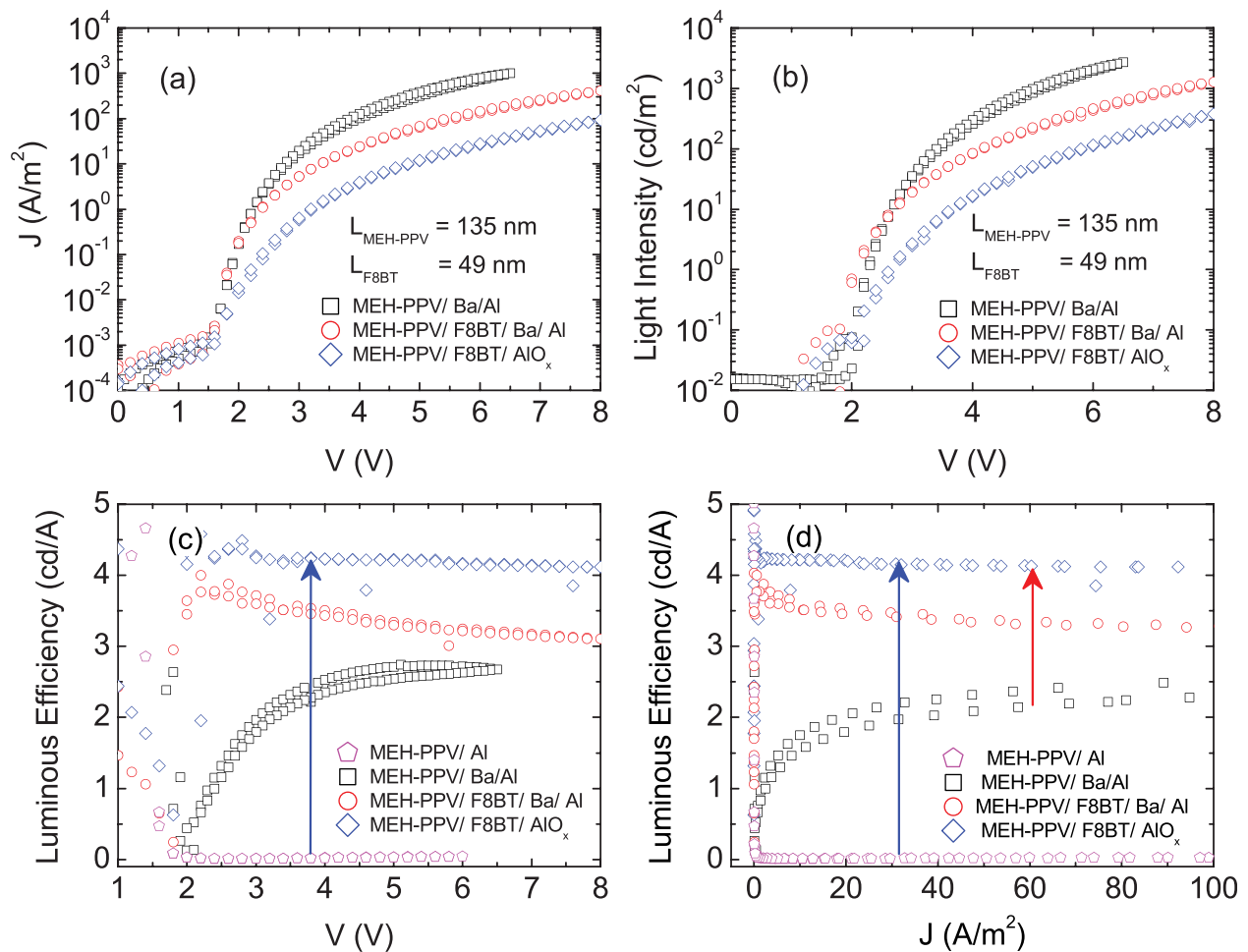


FIG. 5. (a) Current density vs. voltage of single-layer and bilayer PLEDs with both Ba/Al and partially oxidized Al cathodes, (b) light intensity vs. voltage, (c) luminous efficiency vs. voltage, (d) luminous efficiency vs. device current. The blue arrows indicate the efficiency enhancement for a bilayer device with an AlO_x cathode with respect to a single layer PLED with an Al cathode. The red arrow compares the efficiency of a conventional single layer PLED with Ba/Al cathode with a bilayer device having an AlO_x cathode.

bimolecular Langevin recombination, which depends on the product of the hole and electron densities, as compared to the nonradiative trap-assisted recombination, which depends only on the hole density. As a result of the enhanced Langevin recombination, the efficiency of the bilayer device will increase. Due to the absence of exciton quenching and the dominance of Langevin recombination over trap-assisted recombination, the PLED efficiency will be independent of the applied voltage,¹⁹ as observed for the bilayer PLED with AlO_x cathode. For the bilayer PLED with Ba/Al cathode, we observe a slightly lower efficiency and also an efficiency roll-off at higher voltages. Such an efficiency roll-off for PLEDs is typically characteristic for quenching of excitons at the anode.²⁰ We speculate that the better electron injection for Ba/Al as observed in Fig. 4(b) might still lead to an excess of electrons at the MEH-PPV/F8BT interface, such that more electrons can escape the recombination region. For AlO_x the charge balance would be better. An important question is whether an AlO_x cathode will also work for PLEDs other than MEH-PPV, which has a low charge carrier mobility of only $3\text{--}5 \times 10^{-11} \text{ m}^2/\text{Vs}$. Typically, for polymer with a higher mobility the properties of the contact become more critical. Fig. 1 shows that for a single layer PLED of MEH-PPV the AlO_x cathode does not work well. The problem is the large energy off-set between the AlO_x work function (-3.7 to -3.5 eV) and the LUMO of MEH-PPV (-2.9 eV). For F8BT the electron mobility amounts to $3 \times 10^{-10} \text{ m}^2/\text{Vs}$,²¹ so an order of magnitude higher than the electron mobility of MEH-PPV. In spite of this higher mobility the AlO_x cathode works much better, as shown in Fig. 5. The reason is that the energy offset between AlO_x and F8BT (-3.3 eV) is much lower. Another effect that plays a role is the fact that the hole-blocking by the F8BT layer increases the electric field at the injecting cathode. So the question whether an AlO_x cathode will work on other polymers as well depends

on a complex interplay between AlO_x work function, LUMO position of the polymer, electron mobility of the polymer, and field enhancement at the cathode due to hole accumulation.

As stated before, the major advantage of using an Al or AlO_x cathode is their stability in ambient conditions. Devices with Ba/Al cathodes start to degrade already within a few minutes in ambient atmosphere after fabrication. The degradation starts with dark spot growth and within a few hours the PLED does no longer emit light. Figs. 6(a) and 6(b) show pictures of a single-layer PLED with a Ba/Al cathode directly after fabrication 6(a) and after 17 h in ambient atmosphere 6(b), respectively. It is clear that after 17 h in ambient the PLED almost completely turned black. On the other hand, Figs. 6(c) and 6(d) show a bilayer PLED with AlO_x cathode in ambient atmosphere just after fabrication 6(c) and after 30 days in ambient atmosphere 6(d), so *without* encapsulation. We observe that the PLED still uniformly generates light, which demonstrates the superior air-stability of the AlO_x cathode.

IV. CONCLUSION

To summarize, we have fabricated efficient PLEDs with an air-stable AlO_x cathode that still functions after one month in ambient atmosphere. To improve the electron injection and efficiency for AlO_x cathodes, it is necessary to insert an electron transport layer that also blocks the holes from the emissive layer. The accumulation of holes at the interface causes an enhancement of emissive Langevin recombination, leading to an increased PLED efficiency that is independent of the applied voltage and current.

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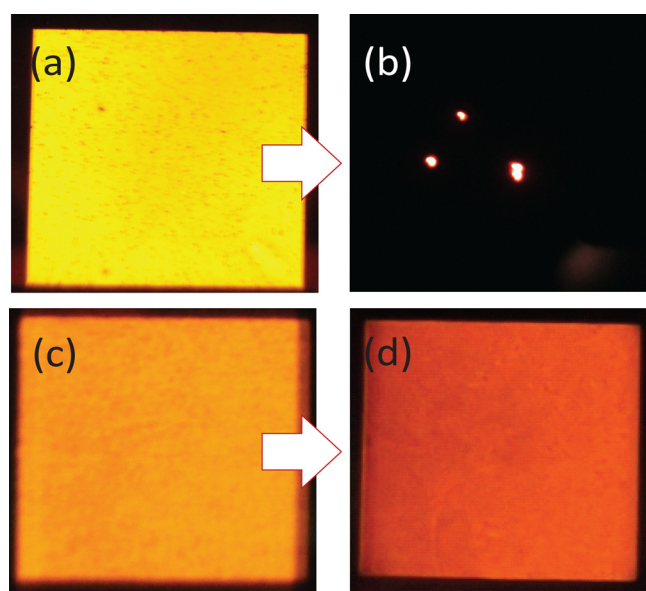


FIG. 6. Pictures from devices ($1 \text{ cm} \times 1 \text{ cm}$) taken at atmospheric ambient, (a) and (b) the single-layer (SL) PLED with Ba/Al cathode after fabrication and the same device after 17 h in ambient, (c) and (d) a bilayer (BL) PLED with AlO_x cathode after fabrication and after 30 days in ambient conditions.

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