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Design of High-Performance Organic Light-Emitting Transistors

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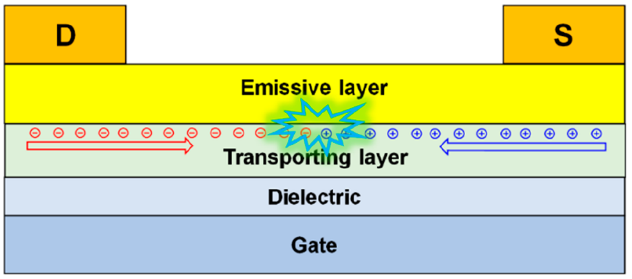
ABSTRACT: Organic light-emitting transistors (OLETs) integrate the light-emitting and gate-modulated electrical switching functions in a single device. Over the past decades, progress has been made in developing new fluorescent semiconductors and device engineering that pushed efficiencies of OLET devices to 8%. However, this efficiency of transistors is still too low to be competitive with organic light-emitting diodes (OLEDs). Currently, there are relatively few suitable organic fluorescent semiconductors suitable for OLETs, and the mechanism of electroluminescence in the devices is still not fully understood. In this mini-review, we discuss the state of highly efficient OLETs and plausible approaches to those unsettled problems. Since this is a mini-review, we will not be able to cover all the excellent work in the literature. Readers are encouraged to read other excellent reviews published earlier.



1. INTRODUCTION

Organic optoelectronics have experienced tremendous growth in recent decades. New organic semiconducting materials exhibiting unique electrical, optical, and magnetic properties were developed and used to create devices with a variety of functionality. Among them, organic light-emitting diodes (OLEDs) represent one of the most significant advancements in the field that has been successfully commercialized. At the same time, another type of organic electronic component, field-effect transistors (OFETs), is still in the experimental stage. Numerous innovations based on OFETs appeared, which stimulated further development in both material design, our understanding of π-electronic systems, and device engineering.[1] The organic light-emitting transistor (OLET) is a unique type of electronic device that combines the advantages of electrical switching from field-effect transistors (FET) and light-emitting property from LEDs. These devices exhibit the potential to simplify the fabrication of existing display systems and may significantly boost progress in new areas, such as photonic communications and electrically pumped organic lasers.[2−5] In an OLED device, electrons and holes are injected into a thin layer of emissive material with a thickness around 100 nm, sandwiched between the two electrodes. At the same time, the electrons and holes in OLET devices are injected through the source-drain electrodes and recombine in the transport channel that is 10−100 μm long. This difference made the development of OLETs more challenge than that of OLEDs.

Ideal light-emitting semiconductors for OLETs need to have appropriate energy levels to minimize charge injection barriers from metal contacts, and they require high, balanced, and, ideally, ambipolar charge carrier mobility to ensure light emission at the center of the channel. They also need to have high fluorescence quantum yield. Therefore, the suitable separation of charge transport and luminescent sites as shown in copolymer polyfluorene[6] is important in designing OLET materials. To achieve efficient exciton to photon conversion and lower operating voltages, a dielectric layer with high k[7] must be used. Design strategies developed in OLED devices such as microcavities, nanoscale structures, patterning of photonic crystals, etc., should be implemented in OLETs. All these parameters must be carefully fine-tuned to balance optoelectronic processes in the device and achieve the most efficient exciton-to-photon conversion. However, one of the challenges of OLET optimization is the fact that these requirements are often mutually exclusive. For example, materials demonstrating high charge carrier mobility usually exhibit efficient $\pi-\pi$ stacking and good electronic coupling to ensure efficient intermolecular charge transport. At the same time, efficient $\pi-\pi$ stacking causes fluorescence quenching due to the formation of exciplexes/excimers, charge transfer states, and other nonradiative decay processes.

High-performance FET semiconductors generally have high charge mobility but low photoluminescence quantum yield (PLQY). For example, rubrene has high mobility of 15 cm^2 V^{-1} s^{-1} due to its tight intermolecular $\pi-\pi$ stacking; however, its PLQY is very low, <1%.[8] In contrast, OLED materials show high quantum yield in the solid state, while the SCLC charge mobility is relatively low (<10^{-3} cm^2 V^{-1} s^{-1}).[9] Thus, mobility fluorescence trade-off is one of the major barriers limiting material design for highly efficient OLETs.[10,11] Several approaches to overcoming this trade-off have been reported

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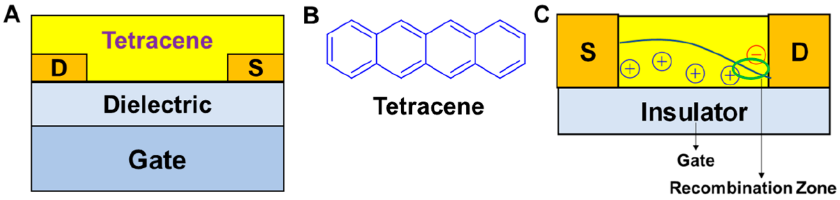

Figure 1. First OLET device based on tetracene. (A) BG−BC FET device structure. (B) Chemical structure of tetracene. (C) The possible mechanism for electroluminescence.

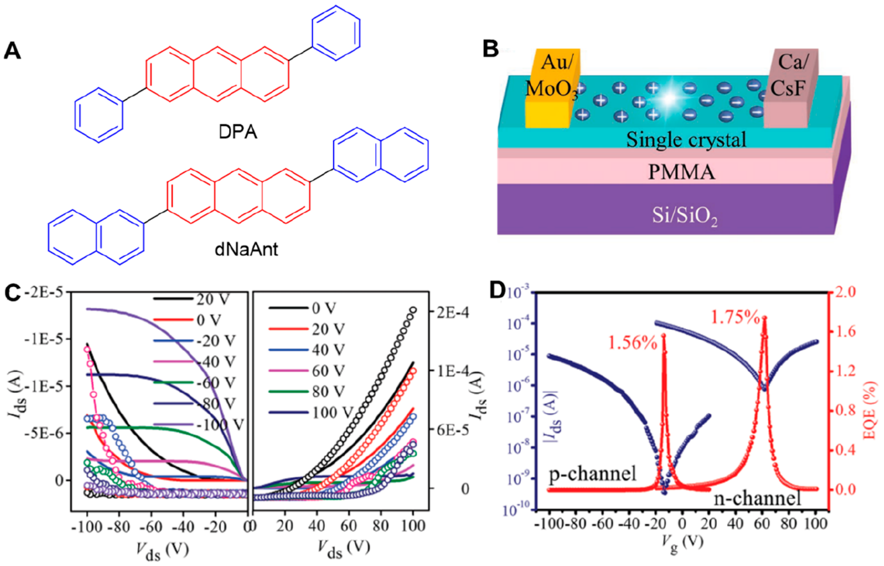

Figure 2. Single-layer single-crystal OLET. (A) Molecular structures of DPA and dNaAnt. (B) Bottom gate−top contact OLET device structure with asymmetric electrodes: hole injected from Au/MoO_3 and electron injected from Ca/CsF. (C) Characteristic FET output curves and (D) transfer curves and EQE change with gate voltage of dNaAnt. Reprinted with permission from the work of Hu et al. Copyright 2019. WILEY-VCH.[14]

in the literature, such as insertion of single-crystal active layers in single-layer devices and multilayer device architectures, where charge transport and light-emitting functions are separated in different layers. In this mini-review we discuss these approaches in more detail.

2. DESIGN OF OLETS

Single-layer OLETs. In 2003, Hepp and co-workers used the bottom gate bottom contact (BG−BC) structure to fabricate single-layer FET devices with an organic small-molecule semiconductor, tetracene (Figure 1A and 1B). The FET devices showed unipolar *p*-type behavior with the hole mobility of 5×10^{-2} cm^2 V^{-1} s^{-1}. For the first time, they observed light emission originating near the drain electrode in a working transistor device. The wavelength of the emission spectral maximum was at 540 nm which is consistent with tetracene's photoluminescence spectrum.[12] The intensity of emitted light and the photocurrent increased with the increasing source-drain and gate voltages but appeared saturated at more negative voltages over −60 V. Based on the energy levels of tetracene, the highest occupied molecular orbital (HOMO) (−5.4 eV) is favorable for hole injection. The energy barrier of 2.7 eV between the lowest unoccupied molecular orbital (LUMO) (− 2.4 eV) of tetracene and the work function of gold electrodes (WF = 5.1 eV) hinders electron injection. The authors attributed the electron injection to electrical field-assisted electron tunneling from the drain electrode (Figure 1C). The holes injected from the source electrode and transported through the channel recombined radiatively with the tunneled electrons near the drain electrode, and light emission was observed. Due to the proximity of light emission to the metal drain electrode, significant quenching was observed. As a result of this quenching and the low PLQY of tetracene film, the obtained EQE of OLET devices was only 6.7×10^{-5}%.

Much better results were achieved for OLET devices with tetracene single crystals.[13] Takahashi and co-workers used bottom gate−top contact configuration with PMMA dielectric and silver paste source-drain electrodes to make FET devices with single-crystal tetracene. When device fabrication and characterization were conducted in an inert atmosphere, without air exposure, they observed ambipolar behavior with the hole and electron mobilities of 9.7×10^{-2} and 1.3×10^{-2} cm^2 V^{-1} s^{-1}, respectively. Even though hole mobility improved almost 2-fold, compared to amorphous tetracene films, there was still a significant energy barrier present for electron injection in the device. To address this issue, the authors used an asymmetric gold−magnesium source-drain electrode. The lower work function of magnesium (3.66 eV) decreased the energy barrier to 1.3 eV and facilitated electron injection. As a result, electron mobility increased almost three times, to 3.7×10^{-2} cm^2 V^{-1} s^{-1}. Light emission was observed in devices with both symmetrical and asymmetrical source-drain electrodes. Due to the ambipolar behavior of the device, the position of the light-emitting zone was dependent on source-drain voltage V_{SD} and moved from the Mg drain to the Au source when the

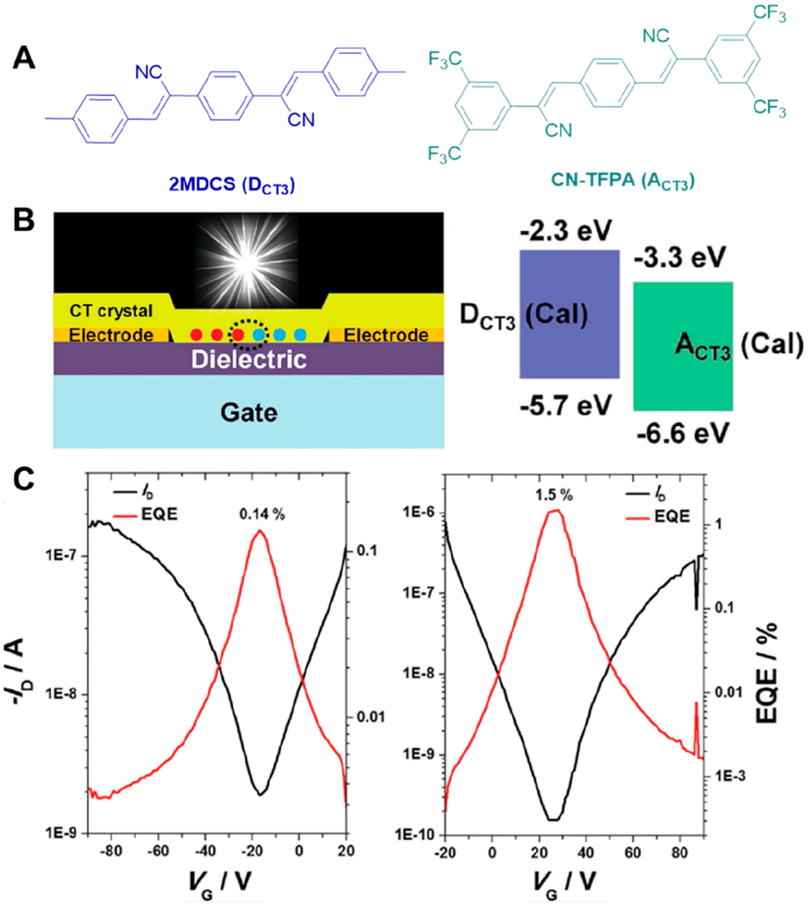

Figure 3. Charge-transfer crystal OLET. (A) Chemical structure of donor, 2MDCS (D_{CT3}), and acceptor, CN-TFPA (A_{CT3}). (B) Schematic illustration and energy level diagram for the BGBC OLET devices. (C) P-channel and n-channel transfer curves and EQE changes with V_G. Reprinted with permission from the work of Park et al. Copyright 2017. WILEY-VCH.[7]

value of V_{SD} decreased from −179 to −255 V. However, no EQE was reported for these devices.

Despite the promising results obtained for the OLET with single-crystal tetracene, the device performance was far from ideal. Unbalanced hole/electron mobility, crystal defects, and low PLQY of tetracene resulted in weak and nonuniform light emission. Many alternative materials have been studied for OLET applications. For example, small-molecule single crystals based on α,ω-bis(biphenylyl)terthiophene (BP3T), 1,4-bis(4-methylstyryl)benzene (CH3-P3 V2), and 2-(4-hexylphenylvinyl)anthracene (HPVAnt) showed both high charge mobility and high solid-state PLQY over 70% which is ideal for OLETs.[3] More recently, Hu and co-workers studied small-molecule materials DPA and dNaAnt as emissive layers for OLETs. DPA and dNaAnt have high-lying LUMOs at around −2.6 eV and high solid-state PLQY of 41.2% and 29.2%, respectively. These molecules with substituted 2,6-positions of anthracene with benzene (DPA) or naphthalene (dNaAnt) moieties (Figure 2A) preserve good photoluminescence properties of anthracene. At the same time, superior charge transport properties are maintained due to extension of π-conjugation length and the formation of J-aggregates in the solid state.[14−16] Both of these materials were used as emissive layers in BG-TC OLET devices with asymmetric source-drain electrodes Ca/CsF (with a low WF of 2.5 eV) and Au/MoO$_3$ (with a high WF of 5.3 eV) (Figure 2B). Both devices exhibited good electronic properties. Devices with DPA achieved hole and electron mobility of 1.99 and 0.33 cm^2 V^{-1} s^{-1}. In comparison, more balanced hole and electron mobilities of 0.73 and 1.11 cm^2 V^{-1} s^{-1} are obtained in dNaAnt, which has more extended conjugated structure (Figure 2C). The OLET devices based on DPA and dNant showed EQE of 1.61 and 1.75%, respectively, which are the highest among single-crystal devices (Figure 2D).

Recently, molecular charge-transfer (CT) cocrystals have attracted research interest in ambipolar charge transport and luminescence.[17] The molecular orbital interactions in the donor and acceptor allow superexchange electronic coupling, thus enabling both hole and electron transport in a single CT crystal. At the same time, high solid-state PLQY is retained, which makes these materials very promising for OLET applications. As demonstrated by Park et al. in 2017,[18] the 2D-type slab CT crystal composed of 2MDCS (D_{CT3}) and CN-TFPA (A_{CT3}) with the ratio of 2:1 (Figure 3A) shows potential as emissive material for OLETs. This cocrystal exhibits high PLQY of 60% in the solid state. The high-lying HOMO (−5.7 eV) in D_{CT3} and low-lying LUMO (−3.3 eV) in A_{CT3} facilitate the injection of both holes and electrons, thus achieving balanced electron and hole mobility of around 10^{-4} cm^2 V^{-1} s^{-1} (Figure 3B). Green emission is observed in a working OLET device with the EQE as high as 0.14 and 1.5% in p-channel and n-channel modes, respectively (Figure 3C).

In addition to small-molecule semiconductors, single-layer OLET devices have also been successfully realized based on polymer semiconductors. In 2006, Henning Sirringhaus and co-workers used poly(9,9-di-*n*-octylfluorene-*alt*-benzothiadia-

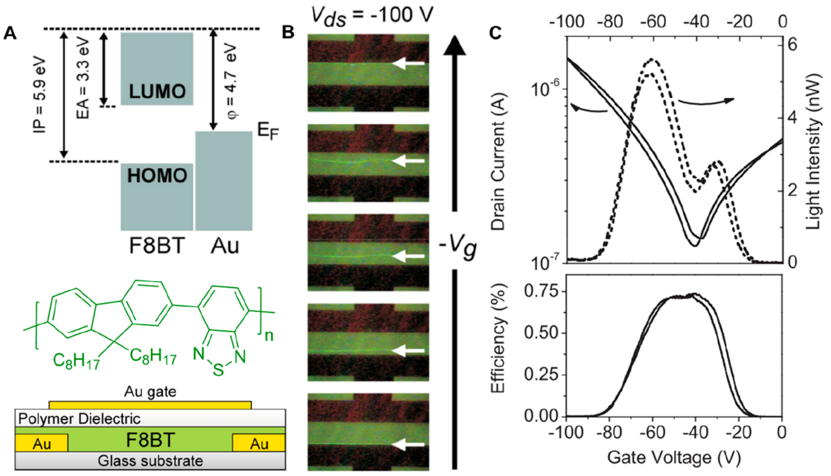

Figure 4. Ambipolar OLET based on F8BT. (A) Energy diagram and top gate−bottom contact (TGBC) device structure for OLETs. (B) Emission zone changes with V_G. (C) Drain current, light intensity, and EQE change with gate voltage. Reprinted with permission from the work of Sirringhaus et al. Copyright 2006. WILEY-VCH.[19]

zole) (F8BT) as an active emissive layer and fabricated top gate−bottom contact (TGBC) OLET devices.[19] A transparent glass was used as a substrate, and source, drain, and gate electrodes were all made of gold. A thick layer of PMMA (400−800 nm) was used as a dielectric and an encapsulating layer. A high-lying HOMO of −5.9 eV and low-lying LUMO of −3.3 eV are found in F8BT, which is favorable for hole and electron injection (Figure 4A). Balanced hole and electron mobilities of 7.5 × 10^{-4} and 8.4 × 10^{-4} cm^2 V^{-1} s^{-1}, respectively, were achieved, as calculated from the transfer curves shown in Figure 4C. In addition, high PLQY of 50−60% is achieved in the solid films, making F8BT a good candidate for OLETs. As shown in the OLET device structure, green emission is found near the electrodes at low gate voltage. The position of the emission zone and light intensity were dependent on the gate voltage. Bright emission from the center of the channel was observed for higher gate voltages. Notably, the highest EQE of 0.75% was achieved in *p*-transport mode (V_{DS} = −100 V; V_G ≈ −40 V). The authors optimized this device structure even further and used silver (Ag) to replace gold as the top gate electrode and inserted zinc oxide (ZnO) as the electron injection layer to form asymmetric electrodes. Much higher EQE of over 8% was achieved in these devices which is a record-performing OLET device to this day.[20] Interestingly, the observed OLET performance is even higher than the corresponding OLED.

2.2. Multilayer OLETs. Due to mobility−photoluminescence trade-off, it is challenging to design highly emissive materials that also exhibit good electrical semiconducting properties. Multilayer OLET devices were introduced as one of the solutions to this problem. Balanced ambipolar charge transport can be achieved by preparing the p−n junction using both p- and n-type fluorescent semiconductors in a single device. The electroluminescent property can then be fine-tuned. For example, by tuning of the coevaporation ratio of p-type semiconductor, a-quinquethiophene (a-5T) and *n*-type semiconductor N, and *N*′-ditridecylperylene 3,4,9,10-tetracarboxylic diimide (P13) in the bulk heterojunction films, Loi and co-workers achieved a large range of control in both charge mobility and electroluminescent intensity.[21] The drawback of this approach is that though charge recombination probability is increased in the bulk heterojunction structure, the flow of charge carrier is impeded, which limits the EL efficiency. In 2010, Sirringhaus and co-workers proposed a lateral heterostructure, in which the tunable recombination zone and high ambipolar charge mobility were achieved.[22] F8BT and poly(9,9-di-*n*-octylfluorene-*alt*-(1,4-phenylene-((4-*s*-butylphenyl)imino)-1,4-phenylene)) (TFB) were deposited through the photolithographic patterning method. Notably, minimal material degradation and high EQE of 1.2% were achieved in these OLET devices.

Multilayer devices have charge injecting, charge transporting, and emissive functions separated in different layers which allows a separate optimization of each component (Figure 5). As shown in Figure 5A, Bazan and co-workers selected a series of fluorescent polymers such as super yellow (SY), poly[2-methoxy5-(2′-ethyhexyloxy-1,4-phenylenevinylene)] (MEH-PPV), and poly[9,9-di(ethylhexyl)fluorene] (PFO) as the emissive layers and designed multilayer OLET devices.[23] In order to facilitate electron injection, a conjugated polyelectrolyte, poly[9′,9-bis[6′(*N*,*N*,*N*-trimethylammonium)-

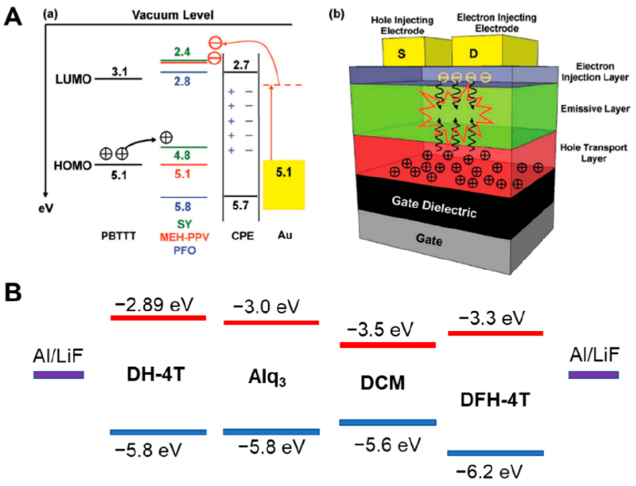

Figure 5. Multilayer OLET. (A) Energy diagram, device structure, and mechanism of electroluminescence in multilayer devices with electron injection layer/emissive layer/hole transporting layer. Reprinted with permission from the work of Bazan et al. Copyright 2011, Wiley-VCH.[23] (B) Energy diagram in the trilayer OLET.

hexyl] florene-*alt*-co-1,4-phenylene], with tetrakis (imidazoyl) borate counterion ($PFN^+BIm_4^-$) was inserted between the source-drain electrodes and the emissive layer. The migration of ions and the dipole effectively modified the interfacial energy and decreased the energy barrier for electron injection. Besides, to ensure good charge transport in OLET devices, a hole-transporting layer poly(2,5-bis(3alkylthiophene-2-yl)-thieno[3,2-*b*]thiophene) (PBTTT) was inserted between the emissive layer and gate dielectric as shown in Figure 5A. As a result of this device engineering, the hole can be directly injected into the HOMO of PBTTT and transported to the vicinity of the drain electrode, where electron is injected to the LUMO of the emissive layer through the electron-injecting layer, $PFN^+BIm_4^-$. Electrons and holes recombine radiatively near the drain electrode, and light emission is observed. All three fluorescent polymers with the different energy levels showed bright electroluminescence and similar EQE of 10^{-3}%. It should be noted that symmetric source-drain electrodes were used in these devices, and all three layers were solution processed. The relatively low EQE is attributed to the proximity of the recombination zone to the metal electrodes due to unipolar charge transport behavior.

Trilayer OLET structures were also successfully realized in thermally evaporated devices, as shown in Figure 5B.[24] 5,5′″-Dihexyl-2,2′:5′,2″:5″,2′″-quaterthiophene (DH-4T) and fluorine-substituted DFH-4T were used as a hole-transporting layer and electron-transporting layer, respectively. The transporting layers exhibited good match of energy levels with the emissive layer material and balanced charge mobility (μ_h = 0.01 $cm^2\ V^{-1}\ s^{-1}$, μ_e = 0.01 $cm^2\ V^{-1}\ s^{-1}$). The emissive layer was composed of the host tris(8-hydroxyquinolinato)-aluminum (Alq_3) and guest 4-(dicyanomethylene)-2-methyl-6-(*p*-dimethylaminostyryl)-4*H*-pyran (DCM). As a result of balanced charge mobility in the device, the observed emission zone was pushed away from the metal electrodes to the middle of the channel, which decreased exciton quenching and resulted in EQE as high as 5% at gate voltage of 30 V. The efficiency obtained in this OLET is nearly 100 times higher than the corresponding OLED, demonstrating the great competitive potential of the OLET.

Besides, different light emission mechanisms were utilized in developing OLETs. For example, Liu et al. utilized the thermally activated delayed fluorescence (TADF) emitters to prepare heterostructure OLETs to harvest the triplet excitons.[25] They developed an exciplex TADF emitter based on *m*-MTDATA (4,4′,4″-tris(*N*-3-methylphenyl-*N*-phenylamino)triphenylamine) as the donor and OXD-7 (1,3-bis[2-(4-*tert*-butylphenyl)-1,3,4-oxadiazo-5-yl]benzene) as the acceptor, and an exciton utilization efficiency of 74.3% for the devices was achieved. Notably, a maximum EQE of 3.76% was achieved in this multilayer OLET device.

3. CONCLUSION AND OUTLOOK

In the past two decades, modest progress has been made in designing high mobility fluorescent semiconductors and optimizing device structures for OLETs. However, there are still several critical issues that need to be addressed: (1) the relative scarcity of high-performance organic materials, (2) low EQE in comparison with OLEDs (>20%), and (3) unclear electroluminescence mechanism in the OLET, especially in the case of multilayer ambipolar devices. In the future work, the introduction of thermally activated delayed fluorescence (TADF) materials and radical emitters[26] in OLET will make a full use of spin dynamics and may overcome the limitations of traditional organic luminescent materials. Moreover, hybrid devices utilizing high mobility inorganic semiconductors, such as carbon nanotube (CNT), graphene, or perovskite, as the transporting layer may become a viable alternative and improve the performance of OLET devices.[27] Meanwhile, development of more efficient dielectrics with high dielectric constant may lower the electrical and optical threshold voltages and realize efficient low-power devices.[7,28] Notably, the integration of additional functions in OLETs may also be an interesting new direction. Recently demonstrated optically switchable OLETs show great promise in device patterning.[29]

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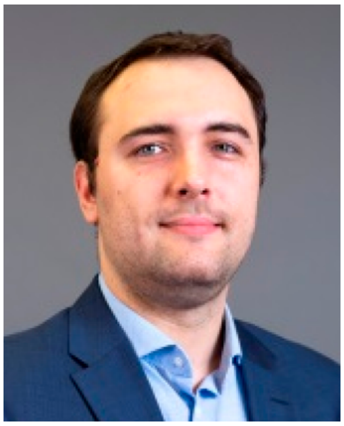

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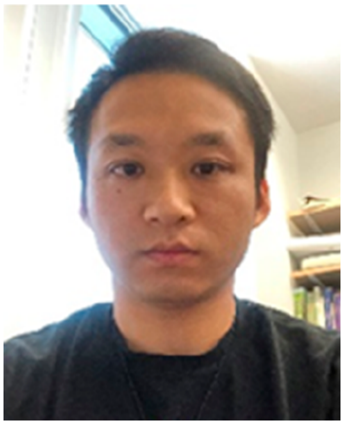

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