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Organic Light-Emitting Transistors Entering a New Development Stage

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Organic light-emitting transistors (OLETs) are possibly the smallest integrated optoelectronic devices that combine the switching and amplification mechanisms of organic field-effect transistors (OFETs) and the electroluminescent characteristic of organic light-emitting diodes (OLEDs). Such a unique architecture of OLETs makes them ideal for developing the next-generation display technology and electrically pumped lasers for miniaturized photonic devices and circuits. However, the development of OLETs has been slow. Recently, some exciting progress has been made with breakthroughs in high mobility emissive organic semiconductors, construction of high-performance OLETs, and fabrication of novel multifunctional OLETs. This recent slew of advances may represent the advent of a new development stage of OLETs and their related devices and circuits. In this paper, a detailed review of these fantastic advances is presented, with a special focus on the key points for developing high-performance OLETs. Finally, a brief conclusion is provided with a discussion on the challenges and future perspectives in this field.

1. Introduction

Multifunctional devices based on organic materials have become important in the field of organic optoelectronics owing to the multifunctionality, low cost, easy processing techniques, and flexibility of the organic materials. As a result, some interesting results have been achieved in the field of organic optoelectronics. The exciting developments in high mobility organic semiconductors, broad-spectrum absorption organic materials, and organic luminescent materials have allowed the construction of high-performance organic optoelectronic devices, such as organic field-effect transistors (OFETs),[1–3] organic photovoltaics (OPVs),[4–6] and organic light-emitting diodes (OLEDs).[7–9] Under the joint efforts of researchers from chemistry, physics, and microelectronics, the development of these devices is progressing steadily. Some of these devices have been commercialized already (such as OLEDs) or are in the process of commercialization. The research on integrated and multifunctional optoelectronic devices have gained momentum owing to the increasing requirements of miniaturized and intelligent devices.[10–13]

Organic light-emitting transistors (OLETs, **Figure 1**a illustrates the typical structure of this device) are possibly the smallest integrated optoelectronic devices that combine the switching and amplification mechanisms of the OFETs and the electroluminescent characteristic of the OLEDs. This unique mechanism makes OLETs a highly integrated device that can emit intense light under gate voltage amplification. The OLETs are considered ideal for developing the next-generation flexible display technology, electrically pumped organic lasers, and optical communication technology.[14–17] Moreover, OLETs also provide a good platform for conducting fundamental studies on charge injection, charge transport, charge carrier recombination, and light emission in organic semiconductors, because of their light-emitting characteristics that can be observed directly.[18,19]

However, following their initial demonstration in 2003 as novel electro-optical devices,[14] the development of OLETs has remained severely impeded and straggled as compared to that of the other organic optoelectronic devices. The OLETs have not been explored extensively by academia and industry.[15–21] For instance, until now, only around 270 papers related to the OLETs have been published, with no more than 25 papers each year (Figure 1b). These studies have been performed mainly by several representative research groups that are concentrated in a few countries around the world. The main reasons for the impeded development of the OLETs are: i) the limited availability of ideal active materials for the OLETs and ii) incompatibility of the conventional device fabrication technology with the OLET structure fabrication requirements.

Recently, some exciting achievements have been reported in this field leading to an upsurge in their research. For instance,

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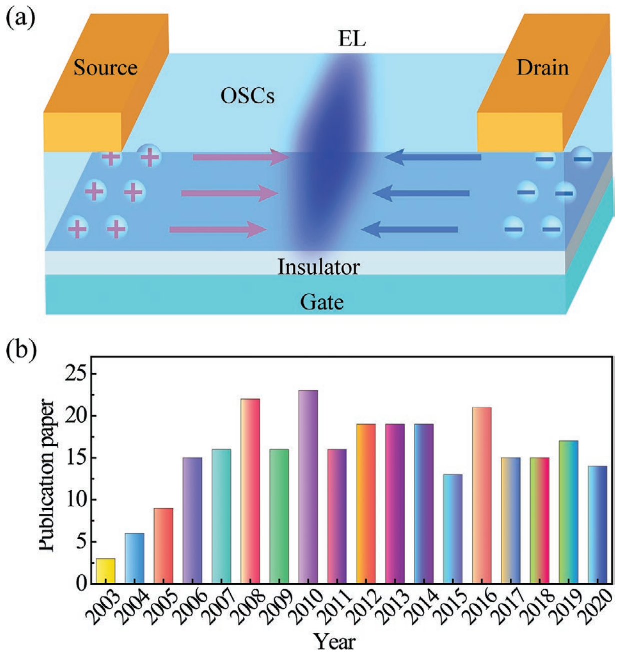

Figure 1. a) Schematic of the device structure and working principle of a typical planar OLET with bottom-gate/top-contact configuration. b) Summary of the number of articles related to the OLETs published from the year 2003 to 2020. The data were obtained from the Web of Science. (OSCs = organic semiconductors and EL = electroluminescence).

with the advancements in chemical and material sciences, breakthroughs have been witnessed for high mobility emissive organic semiconductors, which are the ideal active materials for OLETs, even though the synthesis of high mobility emissive organic semiconductors has been a tremendous challenge for a long time.[22–28] Following the fabrication methods of the currently available high-efficiency OLEDs, various approaches have been applied to the construction of high-efficiency OLETs by balancing the hole and electron charge transport, enhancing exciton utilization, and improving the light outcoupling.[29,30] In addition, some new function-oriented designs of the OLETs have also been proposed to further extend their applications.[31,32] These achievements in the field of OLETs along with the increased interest from academia[33,34] may indicate the advent of a new stage of OLET technology development. This paper is structured as follows: First, based on the basic working process of the OLETs, some key points are proposed for developing high-efficiency OLETs from the perspective of both material sciences and device physics. The current advances in the design and selection criteria of the active layer materials in OLETs are described with the aid of some representative examples. Next, an outline of the potential applications of high-performance OLETs is provided, followed by an overview of the corresponding important advances in different fields. Finally, the challenges and future perspectives of the OLETs are briefly discussed in the conclusion section.

2. Key Points for High-Performance OLETs

As mentioned before, the OLETs are integrated optoelectronic devices that combine the functions of both OFETs and OLEDs. Thus, the factors that affect the charge transport of the OFETs and electroluminescence properties of the OLEDs may also influence the performance of the OLETs. The working mechanism of OLETs can be divided into the following parts: i) charges (both holes and electrons) are injected from the drain and source electrodes separately and transported to the active layers; ii) in the ambipolar region, holes and electrons recombine to form excitons; iii) these high-energy excitons decay to the ground state by emitting photons; and iv) the emitted light is eventually detected as the final output. Similar to the OFETs, high charge-carrier mobility with a good gate modulation capacity is important for improving and controlling the electroluminescent characteristics of the OLETs. Moreover, the OLETs should also have a high $I_{\mathrm{on/off}}$ ratio, low threshold voltage (V_{th}), and small subthreshold slope (SS), which are also crucial parameters. These parameters are determined by the insulator layer, injection barrier, and device structure of the OLETs. In addition, in terms of the electroluminescence, similar to the OLEDs, balanced carrier transportation, and efficient recombination are the other important factors for ensuring sufficient exciton generation, and thus high electro-optical efficiency in the OLETs. The excitons that can be used for electroluminescence depend on the type of luminescent materials, including their exciton utilization and photoluminescence quantum yield (PLQY). A highly efficient light outcoupling and low optical energy loss, which are related to both the orientation of the molecular dipole and device structure, are also critical for developing high-efficiency OLETs.[35] Although some fundamental issues still emerge during the construction of high-performance OLETs, the currently available techniques for the fabrication of OFETs and OLEDs in material sciences and device physics would, to some extent, provide valuable guidelines for the rapid development of OLETs. **Figure 2** shows a schematic representation of these key points for developing high-performance OLETs. These points have been further described in detail in Subsections 2.1–2.2 through some typical examples.

2.1. Active Layer Engineering: High Carrier Mobility and Strong Emission

As mentioned before, the core active layer in OLETs should simultaneously possess high charge-carrier mobility and a strong emission characteristic along with a balanced hole and electron transportation and high exciton utilization abilities. However, in contrast to the OFET- and OLED-based single functional devices, the OLET incorporates optoelectronic functions in the same architecture, which creates special demands and challenges for suitable active materials to be used in the OLETs. To mitigate these drawbacks and achieve the requisite characteristics of an ideal OLET core active layer, single-component, and multilayer device structures have been constructed. Based on the different device structures, different molecular designs, and selection requirements for the active materials are proposed, which have been discussed in detail in Subsections 2.1.1 and 2.1.2.

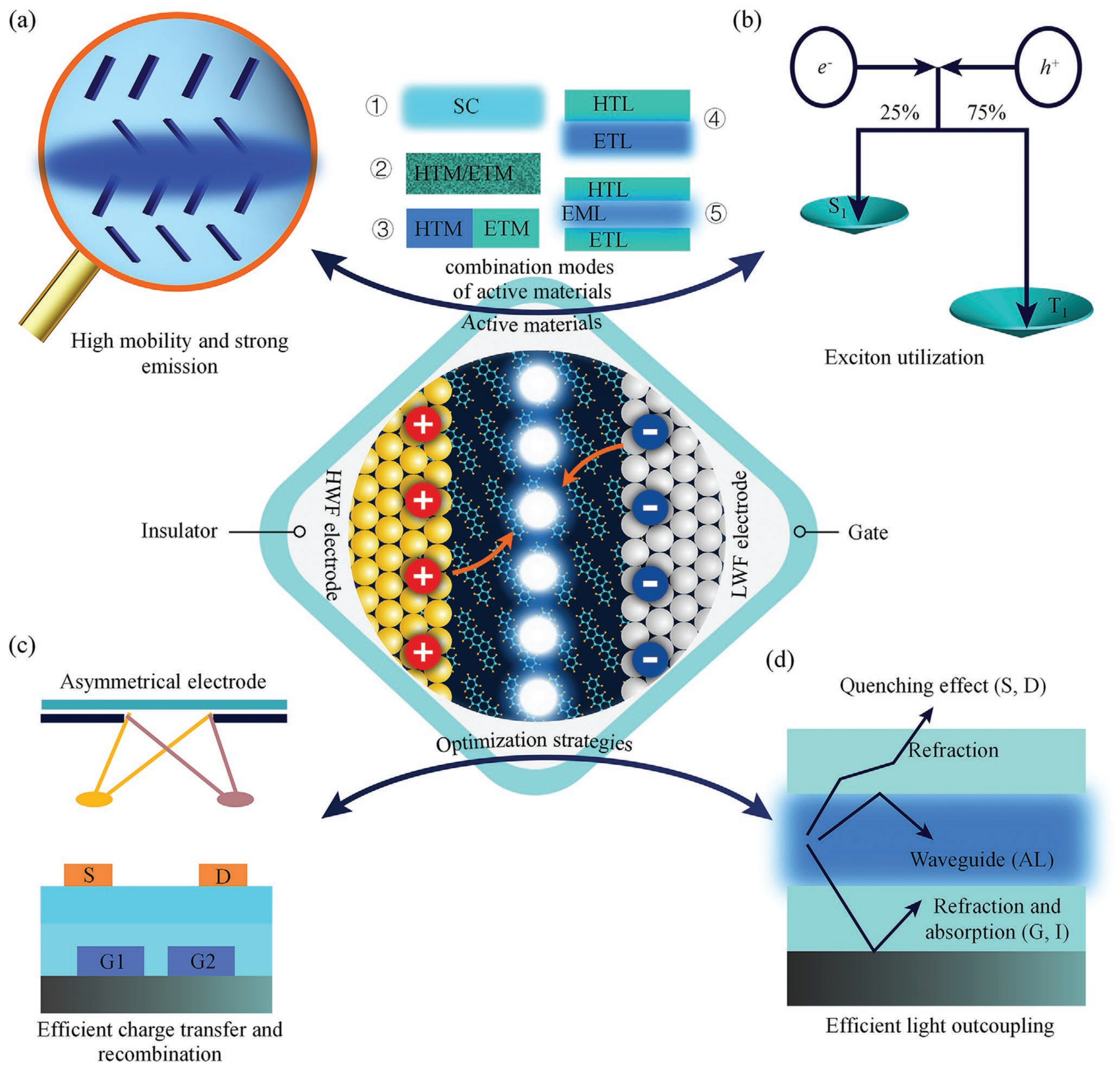

Figure 2. Outline of some key points for the development of high-performance OLETs: a) high mobility and strong emission; b) exciton utilization; c) efficient charge transfer and recombination; d) efficient light outcoupling. (Combination modes of active materials: ① = single component; ② = bulk heterojunction; ③ = lateral heterojunction; ④ = layer heterojunction; and ⑤ = trilayer structure. SC = single component; HTL = hole transport layer; ETL = electron transport layer; HTM = hole transport material; ETM = electron transport material; EML = emitting layer; S = source; D = drain; AL = active layer; G = gate; and I = insulator).

2.1.1. High Mobility Emissive Organic Semiconductors Used in Single-Component OLETs

The advantages of single-layer OLETs are the considerably simplified fabrication technology, low cost, and high integrated density; these are crucial for their real-life applications that include integrated optoelectronic circuits. Such unique working characteristics of the OLETs are also beneficial for studying the charge transportation and recombination properties of organic semiconductors. To achieve these goals, relatively stringent conditions have been proposed for the organic semiconductors, that is, they should simultaneously integrate high charge-carrier mobility and strong emission characteristic in the same layer (as shown in Figure 2a). However, these two properties are mutually exclusive and difficult to integrate, which implies that engineering high mobility emissive organic semiconductors is a challenging task. For example, to construct high mobility devices, organic molecules with extended π-conjugated structures and compact molecular packing are required; in this case, fluorescence quenching will occur and vice versa.[36]

Although some previously reported OLETs that are based on the single-layer structures have demonstrated certain emission properties, the active materials used in these devices generally have high mobility or exhibit strong emission (as shown in **Figure 3**a,b).[37–50] Therefore, the efficiency of such single-layer OLETs is not satisfactory. Fortunately, a series of high mobility emissive organic semiconductors, including thiophene/phenylene co-oligomers, distyrylbenzene derivatives, anthracene derivatives and so on, have been demonstrated (Figure 3c).[51–65] For instance, α, ω-Bis(biphenylyl)terthiophene (BP3T), which is one of the thiophene/phenylene co-oligomers, exhibited a high PLQY of 80% with carrier mobilities of 1.64 and 0.71 $cm^2 V^{-1} s^{-1}$ for the holes and electrons, respectively.[52]

In 2016, Ma and co-workers reported the synthesis of 1,4-bis(2-cyano-2-phenylethenyl)benzene (β-CNDSB) by introducing cyano groups into distyrylbenzene derivatives.[28] The target molecule demonstrated aggregation-induced emission (AIE) characteristic with a PLQY up to 75% in their single-crystal states. The highest mobility of an OLET that is based on β-CNDSB approached 2.10 and 2.50 $cm^2 V^{-1} s^{-1}$ for the holes and electrons, respectively. Since 2015, our group has carried out some investigations in this field and made breakthroughs in designing high mobility emissive anthracene-based derivatives.[22–24,36,66–68] The design is inspired by the molecular structure of strongly emissive anthracene and high-mobility pentacene. To enhance the charge-carrier mobility of anthracene and reduce the fluorescence quenching effect, which is induced by a coplanar π-structure similar to that of pentacene, we introduced a rotatable carbon–carbon single bond between the anthracene core and π-extended moiety for synergetic modulation of the electrical transportation and emission properties. To date, tens of new anthracene-based high mobility emissive organic semiconductors with symmetric and asymmetric structures have been successfully developed with tunable optoelectronic properties.[22–24,36,66–68] Among them, for a single crystal, 2,6-diphenylanthracene (DPA) and 2,6-di(2-naphthyl)anthracene (dNaAnt) have exhibited superior mobility of 34 and 12.3 $cm^2 V^{-1} s^{-1}$ with PLQY up to 41.2% and 29.2%, respectively.[22,23] Such a molecular design has been successfully extended to other high mobility emissive organic semiconductors, such as fluorene-based high mobility organic laser semiconductors, which are important for developing electrically pumped organic lasers.[25] These achievements in the case of high mobility emissive organic semiconductors further promote the research on single-component OLETs.[22,29] Currently, the high mobility emissive organic semiconductors and single-component OLETs have gained considerable attention and interest in material-based research.

Overall speaking, the current research on the high-mobility emissive organic semiconductors is still in the trial-and-error stage, lacking rational design guidelines, and an in-depth theoretical understanding of the intrinsic properties of the materials. There is a consensus that the incorporation and modulation of appropriate luminescence centers and π-extended units are crucial for the integration of high mobility and strong emission characteristics. While trying to increase the π-conjugation of backbones with compact intermolecular packing, the resulting significant quenching effect should be avoided; thus, a subtle balance between these two properties is needed to maximize their integration. Considering this, the construction of a certain torsion molecular structure and the incorporation of large side chains in the molecules are two effective approaches that are used currently.[24,36,69–72] Moreover, a deep understanding of the effect of molecular structures and intermolecular interactions on the resulting electrical and optical properties[1,73–75] would also provide valuable information. Apart from the improvement of the optoelectronic properties of materials, some other parameters should also be considered for achieving enhanced OLET efficiency. For instance, most of these high mobility emissive organic semiconductors are mainly fluorescent materials. In fluorescent materials, 25% of the total excitons are available for electroluminescence. Improving exciton utilization is an effective approach for significantly improving the efficiency of the OLETs, which are similar to the OLEDs (as shown in Figure 2b). Thus, the development of high mobility phosphorescent, triplet–triplet annihilation (TTA),[76] and thermally activated delayed fluorescence (TADF) materials is necessary for further advancements in the OLET technology. Additionally, the efficiency of the excitons that release energy via light emission depends on the PLQY of the luminescent material; thus, a higher PLQY results in better efficiency. From the aspect of electroluminescence, along with the high charge transportation properties, balanced carrier transportation and efficient recombination are also crucial for generating sufficient number of excitons. However, the currently available materials are mostly unipolar semiconductors with p-type transportation.[1,24,36] The OLETs that are fabricated from these materials require complex device structures to enhance the electron injection for balanced charge transportation within the active layer. For engineering advanced electrically pumped organic lasers and novel photonic circuits, the development of superior organic laser semiconductors is crucial.[10,25,52,77] Recently, Shuai and co-workers[78] reported a computational screen-out strategy based on a set of previously reported optical pumping organic laser molecules. They pointed out that for the electrically pumped organic laser materials, to avoid the exciton and/or polaron reabsorption and the accumulation of triplet excitons, a large oscillator strength (S_1) and weak intermolecular π–π interaction are preferred. These electrically pumped organic laser materials provide valuable guidelines for further investigations in this field.

2.1.2. Active Materials Used in Multilayer OLETs

Considering the trade-off between the charge transportation and light emission in one organic molecule and the challenge of developing high mobility emissive organic semiconductors, one of the alternative approaches for developing high-performance OLETs is to fabricate multilayer OLETs, similar to the OLEDs. In such devices, the functionalities of the charge transportation and light emission are separated in different layers, such as the charge injection layer, charge transportation layer, and light-emitting layer. Several combination modes of the active materials that are used in multicomponent OLETs have been summarized in Figure 2, such as bulk heterojunction, lateral heterojunction, layer heterojunction, and trilayer structure. These combination modes have various advantages as well as limitations. Next, we will discuss the recent advances

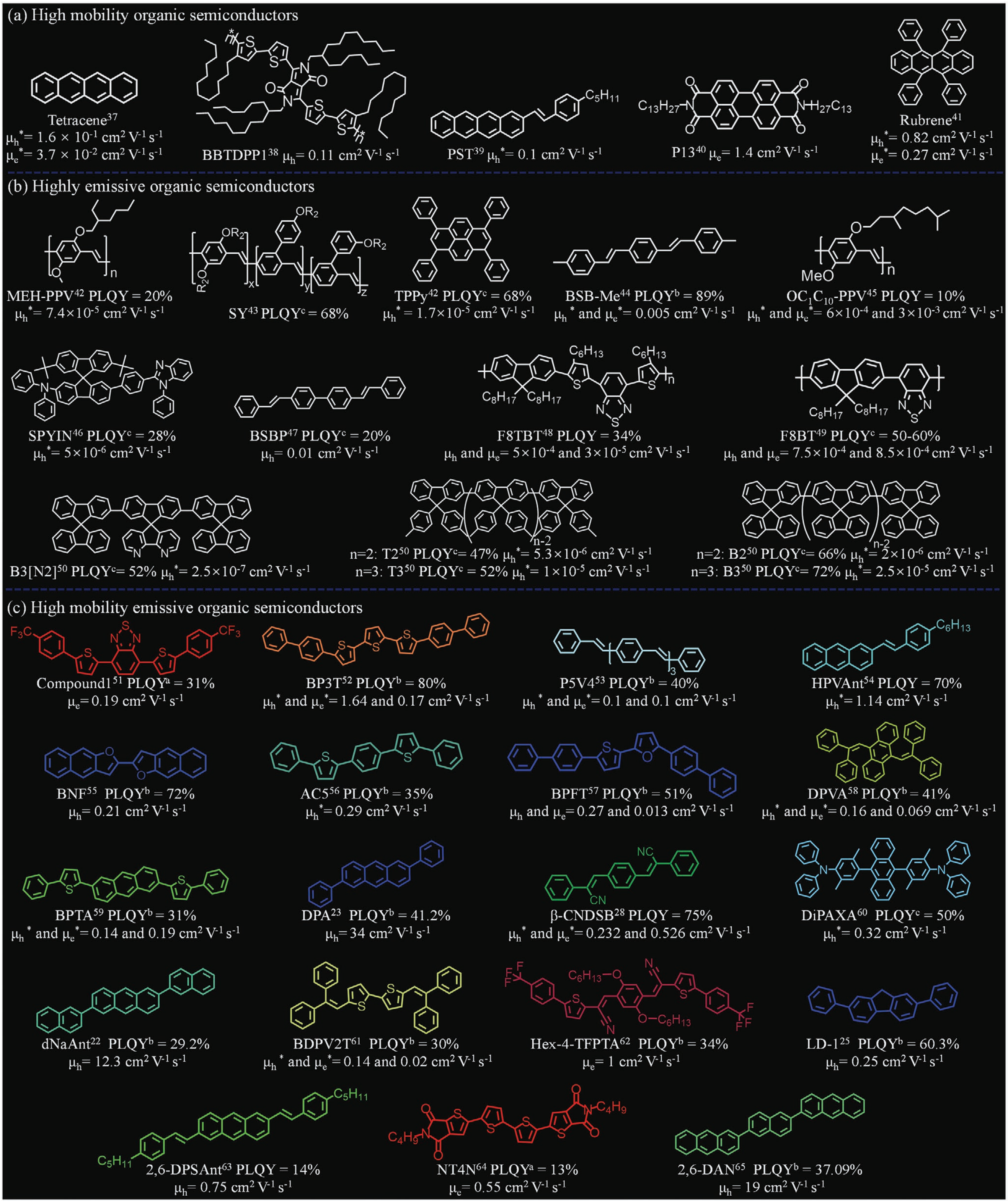

Figure 3. Chemical structures of the a) high mobility organic semiconductors, b) highly emissive organic semiconductors, and c) high mobility emissive organic semiconductors currently used in single-layer OLETs. (a, b, and c represent the test conditions of PLQY in solution, single crystal, and film, respectively. * indicates that mobility is from OLETs, others come from OFETs. The color of the molecular structure represents the electroluminescence color of the molecule. R2 = $OC_{10}H_{21}$).

in active materials that are used in the multicomponent OLETs based on the above-mentioned combination modes. The ideal energy of the level matching diagram of the active materials used in multilayer OLETs is illustrated in **Figure 4**a. For easy electron and hole injection into the light-emitting layer for recombination, the ideal energy level matching will occur when the highest occupied molecular orbital (HOMO) energy level of the luminescent material lies above the HOMO energy level of the hole transport material and the lowest unoccupied molecular orbital (LUMO) level remains below the LUMO level of the electron transport material. At the same time, to reduce the light loss in the process of light outcoupling, the emission spectrum of the luminescent material should not be within the central absorption range of the charge transport materials. In 2004, Muccini et al. reported an OLET based on a bulk heterojunction film of α-quinque-thiophene (α-5T) and *N*, *N*′ditridecylperylene-3,4,9,10-tetracarboxylic diimide (P13) with a ratio of 1:1.[79] The device exhibited balanced charge transportation and controllable electroluminescence with hole and electron mobilities up to 10^{-4} and 10^{-3} cm^2 V^{-1} s^{-1}, respectively. Although this kind of bulk heterojunction that is obtained by co-evaporation can easily achieve ambipolar charge transportation characteristics, to some extent, it may significantly disturb the orderly packing of the molecules, leading to a low device efficiency. In 2006, an OLET based on bilayer heterojunctions of α, ω-dihexyl-quaterthiophene (DH-4T) and P13 was reported by Dinelli et al.[80] By studying the influence of the deposition order on the device performance, the authors reported that the "growth compatibility" between the two materials is conducive for the formation of continuous interfaces and beneficial for achieving a high efficiency of the OLET.

For OLETs based on organic semiconductor heterojunctions, p-type materials are often used as the hole transport layer and n-type materials are used as both the electron transport layer and light-emitting layer. This is because the holes easily enter the HOMO level of n-type materials from the HOMO level of

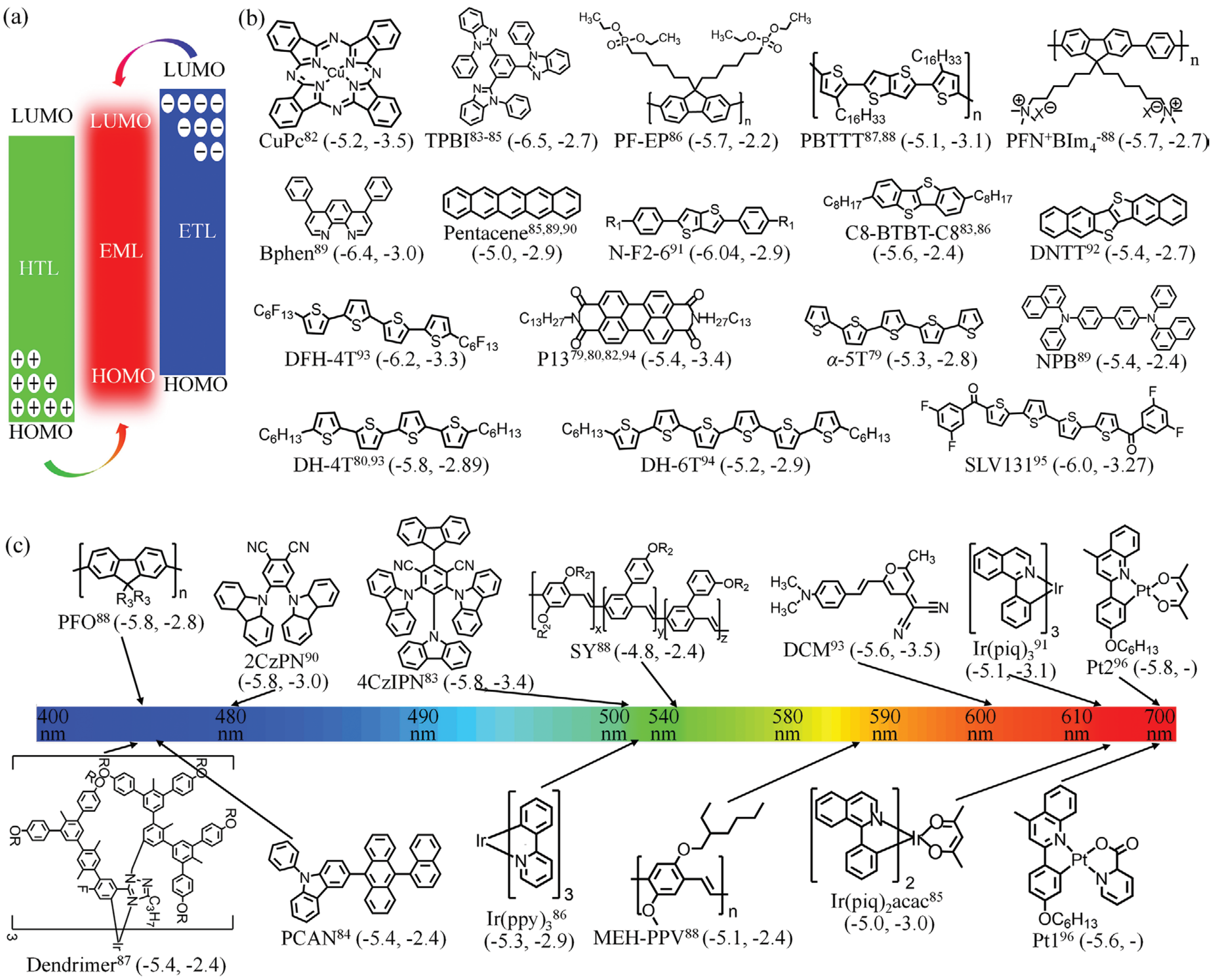

Figure 4. a) Ideal energy of the level matching diagram of active materials used in trilayer OLETs. b) Electrical transportation materials, and c) emissive materials currently used in multilayer OLETs. (The numbers in parentheses after the molecular abbreviation are the HOMO energy level and LUMO energy level of the molecule. R = 2-ethylhexyl, R1 = C_8F_{17}, R2 = $OC_{10}H_{21}$, R3 = ethylhexyl).

p-type materials. The low efficiency of these devices is mainly attributed to the limited availability of the desired organic semiconductor materials with high electron mobility and strong emission. In addition, some bilayer OLETs, reported so far, usually exhibit unbalanced charge transportation with a p-type dominating mode, thereby leading to a relatively low external quantum efficiency (EQE). A trilayer structure is realized by inserting a light-emitting layer between the hole and the electron transport layer. The hole transport layer, electron transport layer, and light-emitting layer are completely separated in the trilayer OLETs, which is considered to be an ideal structure for the development of high-performance OLETs with a balanced charge transportation characteristic. In 2013, Namdas and coworkers successfully constructed a trilayer semiconductor heterostructure based OLET with a nonplanar source/drain contact architecture.[81] The authors emphasized that the OLET with a nonplanar source/drain contact architecture exhibited higher brightness, EQE, and $I_{on/off}$ ratio than the device with planar source-drain contact architecture. This is observed owing to the decrease in contact resistance, which increases the recombination efficiency of the holes and electrons. These results reveal that the bilayer or multilayer structures are promising for the development of high-performance OLETs, which are compatible with the currently investigated OLEDs. Some of these representative charge transport and light-emitting materials are summarized in Figure 4b,c.[82–96] Notably, although the ideal energy levels should be similar to those shown in Figure 4a, in most cases, a certain injection barrier due to the limitation of the optional material system is usually observed. Light loss caused by the absorption of electrical transportation materials has not received sufficient attention. Thus, considerable efforts should be made in this field for developing OLETs with much higher efficiency. In addition, for this part of the research, it is also necessary to refer to the research strategies of high-performance OLEDs and OFETs related to the material selection and optimization approaches.[1,2,9,97]

2.2. Device Structure Engineering: Efficient and Balanced Hole and Electron Transportation, Low Optical Loss, and High Light Outcoupling

In principle, as a unique electroluminescent device, OLET has the capacity to move light emission zones far away from the source/drain electrodes in the planar geometry. This characteristic is beneficial for decreasing the electrode quenching effect and efficiency roll-off, and improving the lifetime, thereby yielding a high EQE and brightness.[15] However, the current efficiency of OLETs is considerably lower than their expected values when benchmarked against OLEDs. To fabricate high-efficiency OLETs, apart from developing ideal active materials, device structure engineering is also crucial. The definition of EQE is as follows: EQE = $\gamma \times \eta_{S/T} \times \eta_{PL} \times \eta_{out}$; where γ is the electron–hole balance, $\eta_{S/T}$ is the singlet-triplet factor (25% for the fluorescent emitters, 62.5% for the TTA emitters, and 100% for the phosphorescent emitters), η_{PL} is the PLQY, and η_{out} is the outcoupling efficiency. The properties of charge transport, $\eta_{S/T,}$ and η_{PL} depend primarily on the active materials. For γ, in the ideal situation, the active materials have intrinsically well-balanced ambipolar charge transport; thus, a value of $\gamma \approx 1$ can be easily achieved based on simple device fabrication. However, currently, most of the high mobility and high mobility emissive organic semiconductor materials are unipolar with p-type dominating transportation properties.[1,24,36] The unbalanced hole and electron charge transport yield a very low value of γ ($\gamma << 1$). To solve this problem, constructing an asymmetric device geometry, especially with the incorporation of a low work function (LWF) electrode is a widely used approach in the OLETs for the improvement of electron injection and transportation (Figure 2c top).[22,29,98,99] One of the challenges in such a device geometry is the instability of the LWF electrode, which increases the complexity of the device fabrication (special inert environments and multistep fabrication are required) and instability of the resulting device performance. Thus, a perfect γ value is difficult to achieve because of the inefficient minority charge carrier (electrons) transports under such a single-gate-modulated structure, which is unfavorable for a large amount of exciton formation in the recombination zone. Another interesting strategy has been developed to address these problems, where a split-gate and overlapping split-gate device is designed with the concept of controlling the holes and electrons, independently, via different gates for a better balance of these two charges and higher excitons density in the recombination zone (Figure 2c below).[92,100] In addition, similar to the OLEDs, multilayer OLETs with separate hole and electron injection layer and transport layer are also used for achieving high efficiency by balancing the holes and electrons in the emitting layer, which we have mentioned in Section 2.1.2

Further, η_{out} is the outcoupling efficiency, which is also an important factor affecting the performance of the resulting device. η_{out} can be significantly enhanced by rationally optimizing the device structure. Owing to the optical losses caused by the surface plasmon polaritons (SPP), waveguide mode in the active layers, substrate absorption mode, and electrode quenching, the η_{out} of OLEDs is usually 20–30%. For the OLETs, the optical transmission path is completely different. In a typical planer OLET with a bottom-gate top-contact configuration, the generated light can be directly out coupled from the active layer, without the requirement of passing through many active layers and electrodes. In addition, owing to the unique structure of the planar OLETs, the emitted light can be well-controlled via the applied gate voltages and moved far away from the source and drain electrodes. Therefore, the optical loss caused by the SPP and interaction between the electrode and excitons can be largely avoided. Considering these points, it is easy to achieve high light-outcoupling efficiency. However, the difference in refractive index (n) between the organic layer and air is quite considerable (0.6–0.8), which leads to a low probability of light outcoupling for the OLETs.[30,35] In this case, optical waveguide losses in the active layer may severely affect the light coupling output efficiency of OLETs. In addition, although the effect of electrode quenching is reduced owing to the light-generating zone is far away from the source and drain electrodes, the optical loss induced by the absorption of the gate electrodes cannot be neglected. This is another important factor that causes high optical losses in the OLETs. Recently, a systematic theoretical calculation has been performed by Lee et al. to analyze the coupling efficiency of multilayer OLETs. Their

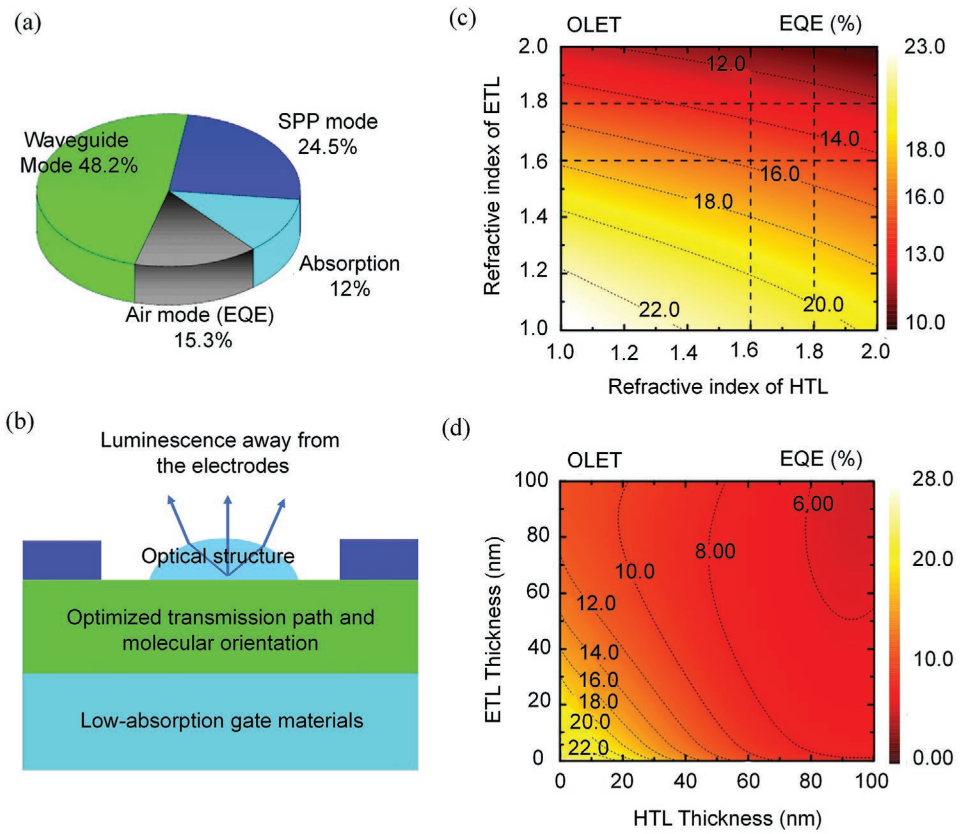

Figure 5. a) Optical coupling losses caused by several modes in the plane OLETs. b) Schematic diagram of reducing optical coupling losses through device structure engineering. Calculation results of EQE versus c) n and d) thickness of the active layer. a,c,d) Reproduced with permission.[35] Copyright 2019, AIP Publishing.

analytical results demonstrate that the main optical energy losses in such OLETs are induced by the waveguide mode in the active layer and absorption of the Si gate (as shown in **Figure 5**a–d).[35] For the OLETs, by modifying the device structure, such as adjusting the thickness and the n of the active materials, changing the gate materials, varying the molecular orientation, and incorporating special optical structures for realizing efficient optical transmission paths, much higher EQE values (even better than those of the OLEDs) can be achieved (Figure 5b). Moreover, in addition to the high EQE, some other special requirements should also be considered, such as the investigation of electrically pumped lasers and electrically driven photonic devices or circuits, which require optimal optical resonators and/or special structures.[10,101]

3. Current Important Advances in the OLETs and Their Applications

As mentioned before, the unique feature of OLETs with integrated functions of OFETs and OLEDs in the same architecture enables them to have potential applications in numerous fields. The OLETs have received widespread attention from academia and industry, mainly because of their inherent self-luminous and self-driving characteristics, which can further simplify the structure and manufacturing process of the prevailing active-matrix organic light-emitting diode (AMOLED). Another reason for the scientific community's attention on the OLETs is their potential use in the construction of electrically pumped lasers, which are a difficult yet significant research subject. In addition, multifunctional devices based on the OLETs, such as up-conversion devices and optically controllable devices, are emerging.

3.1. High-Performance OLETs

Owing to the integrated features of charge transport and luminescence, high mobility emissive materials are the ideal candidates for fabricating high-efficiency single-component OLETs. In Section 2.1.1 a series of recently developed high mobility emissive organic semiconductors have been described. These developments indicate a promising future for research on high-efficiency single-component OLETs. Among them, BP3T, which is a typical integrated optoelectronic material combining the properties of charge transport, strong light emission, and lasing characteristics, has been widely investigated previously because of its applications in organic electronic and optoelectronic devices.[102,103] In 2012, Takenobu et al. fabricated light-emitting transistors based on the BP3T single crystal with high current density and high EQE.[104] To achieve effective electron injection, an electron injection buffer layer (CsF) was introduced to reduce the injection barrier for achieving balanced hole and electron mobilities (both mobilities ≈ 0.2 cm^2 V^{-1} s^{-1}); the resulting EQE value approached 1%. Simultaneously, the current density of the device was further increased by narrowing the channel of the transistor. The resulting current density approached 33 kA cm^{-2} (assuming the current flow to be confined at one molecular layer). Currently, based on the developed high mobility emissive organic semiconductors, a rapid development stage has been reached, especially for the single-component OLETs.[22,29,65] For instance, two superior anthracene-based high mobility emissive organic semiconductors (DPA and dNaAnt) have been used as the active layer for the construction of OLETs. Modulating and enhancing the hole and electron injection and transport of devices have been performed by optimizing the energy level matching. Owing to the intrinsic ambipolar properties, spatially well-controlled and narrowed recombination zone in the conducting channel have been demonstrated in both the p- and n-operation regimes (**Figure 6**a–e).[29,105] Finally, a high EQE value of 1.61% for DPA-OLETs and 1.75% for dNaAnt-OLETs have been obtained by optimizing the device fabrication method and crystal quality. The EQE values of these devices have set a new record for the single-component device efficiencies that have been obtained in the past few years (Figure 6f,g). Hu et al. demonstrated the construction of yellowish-green emissive OLETs based on another highly π-extended anthacene derivative, 2,6-DAN compound, which produced an emission spectrum that was shifted to 700 nm with the strongest peak at around 550 nm.[65] Recently, a cyano-substituted styrene derivative was synthesized and used for the construction of OLETs by Ma et al. Owing to the high-quality single crystal and low optical loss, the EQE of the device reached an unprecedented value of 2.02%.[106] These encouraging results indicate a promising future of constructing high-performance OLETs with the development of high mobility emissive organic semiconductors.

In comparison with the single-component OLETs, the development of multilayer OLETs has been more productive because they can realize the electrical and emission functionalities in different layers, which, to some extent, circumvents the challenge of developing high mobility emissive organic semiconductors. In 2010, Muccini and co-workers reported an OLET based on a p-channel/emitter/n-channel trilayer semiconductor heterostructure, as shown in Figure 6h.[93] DH-4T, di(perfluorohexyl-quaterthiophene (DHF-4T), and a blend of host tris(8-hydroxyquinolinato)aluminum (Alq3) and guest 4-(dicyanomethylene)-2-methyl-6-(p-dimethyl-minostyryl)-4H-pyran (DCM) were used as the hole transport layer, electron transport layer, and light-emitting layer, respectively. For this device, the good matching of the energy levels between the transport materials and emissive materials resulted in an EQE of ≈5% (Figure 6i), which was 100 times more than the EQE of the equivalent OLED. The result was a significant achievement for the OLETs at that time and suggested promising applications of the OLETs in display technology.

Fluorescent materials have been widely used in the investigation of OLETs because of their characteristics of high color

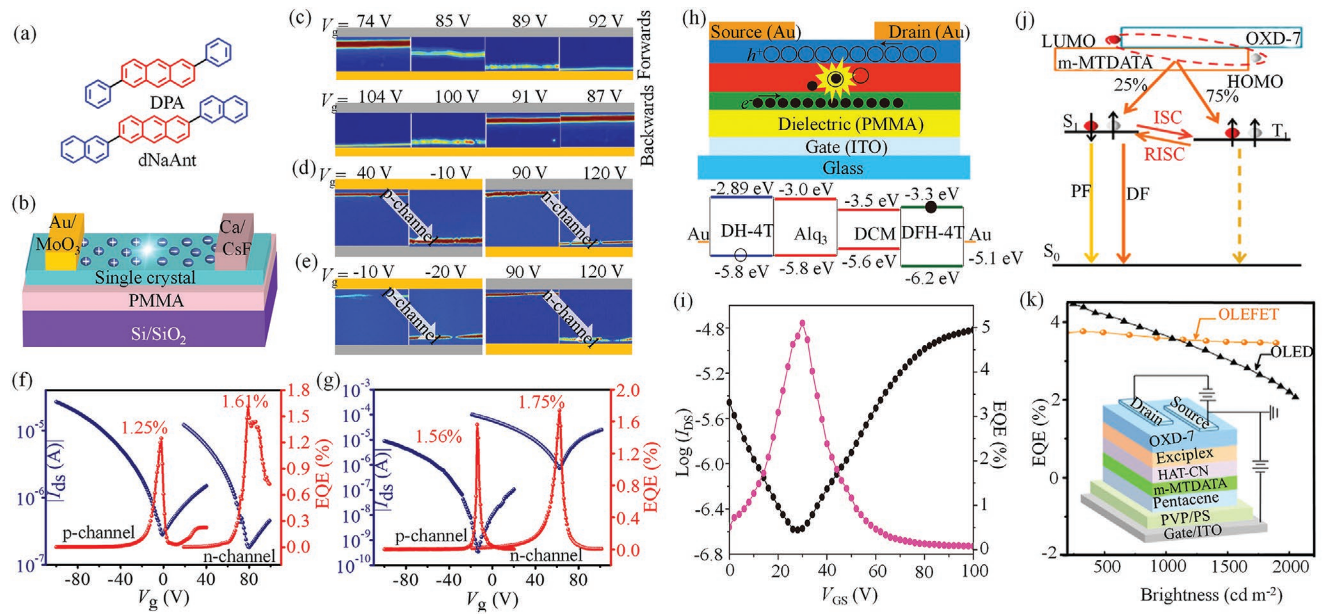

Figure 6. a) Chemical structure of DPA and dNaAnt. b) Schematic of the bottom-gate/top-contact device structure with asymmetric electrodes. c) Color-code images of dNaAnt-OLET extracted from light emission under forward and backward. d,e) Color-coded images of d) DPA–OLET and e) dNaAnt-OLET under p- and n-channel operation. f,g) EQE versus *Vg* for f) DPA–OLET and g) dNaAnt–OLET within both p- and n-channel transfer curves. h) Schematic and energy-level diagram of OLET based on p-channel/emitter/n-channel trilayer semiconducting heterostructure. i) EQE versus V_g for OLET as illustrated in (h) within n-channel regions. j) Schematics of formation of excitons in the luminescent layer of OLET based on exciplex TADF material. k) EQE versus brightness of OLET based on exciplex TADF material. a–g) Reproduced with permission.[29] Copyright 2019, Wiley-VCH. h,i) Reproduced with permission.[93] Copyright 2010, Springer Nature. j,k) Reproduced with permission.[108] Copyright 2017, American Chemical Society.

purities, high PLQY, and long-term stabilities.[25,29,52] However, the major disadvantage of fluorescent materials is that only 25% of the single excitons can be utilized for the electroluminescence process with 75% triplet excitons being wasted, as indicated by the spin-statistics theorem limit. Based on the high exciton utilization luminescent materials that have been used in the OLEDs, phosphorescent materials and TADF materials have been used for the development of high-performance OLETs. In 2009, Samuel and co-workers successfully fabricated bilayer OLETs using phosphorescent materials for the first time and achieved a peak EQE of 0.45% at 480 cd m^{-2}.[107] The phosphorescent material was doped in a suitable charge transport material owing to the low mobility of the phosphorescent materials, which is insufficient for their direct applications in the OLETs. Namdas and co-workers reported OLET based on a host-free blue phosphorescent dendrimer with a brightness of 650 cd m^{-2} and an EQE of 2.1%, which are comparable to those of equivalent OLEDs.[87] Moreover, negligible EQE roll-off in this OLET at high current density and brightness was observed. Recently, Meng et al. demonstrated a phosphorescent material-based OLET exhibiting high-performance with an EQE of 9.01%.[33] In 2017, Liu and co-workers reported an exciplex TADF material-based OLET (Figure 6j). The OLET based on the exciplex of 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 showed a maximum EQE of 3.76% and an $I_{on/off}$ ratio of ≈10^4 (Figure 6k).[108] Another excellent example of OLET based on a TADF material, 4CzIPN (2,4,5,6-tetra[9H-carbazol-9-yl]isophthalonitrile), is also reported. By incorporating a hole-blocking layer in the OLET architecture, an average brightness of over 500 cd m^{-2} with an EQE of 0.1% is achieved.[83] These examples demonstrate that the increase in high exciton utilization is crucial for further improving the performance of OLETs. Although the current EQE values obtained in these devices are still low, it is believed that much higher EQE values would be achieved with further optimization.

Several methods have been reported for improving the outcoupling efficiency of the OLEDs,[109] such as the microlens array (MLA) patterning on the substrates, introduction of a light-scattering layer, application of porous nanocellulose paper, and insertion of a photonic crystal layer. Some techniques have been used to efficiently enhance the outcoupling efficiency of the OLETs, including both external and internal light extraction outcoupling. In 2011, Heeger and co-workers fabricated an optoelectronic gate dielectric consisting of alternating multilayer materials with high and low *n* and used it in an OLET that emitted light from the top.[110] Compared to the SiNx gate dielectric, the optoelectronic gate dielectric can redirect the collected light in the forward direction to improve the brightness and efficiency of the OLETs simultaneously. The efficiency increased by 4.5 times with respect to the previous value, reaching 0.9 cd A^{-1} at the brightness of 4500 cd m^{-2} as compared to the OLET with SiNx gate dielectric. Sirringhaus et al. invented a strategy for enhancing the external light extraction outcoupling in a later report, as shown in **Figure 7**a,b.[30] The bottom-contact/top-gate OLET with bottom emission, based on poly(9,9-di-n-octyl-fluorene-alt- benzothiadiazole (F8BT), was fabricated, and the efficiency of the device was improved to a satisfactory value with a peak EQE greater than 8% by adding a half-sphere in the direction of light emission for enhanced light collection (Figure 7c). At present, limited efforts are being made to improve the outcoupling efficiency of the OLETs. Effective strategies and technology are urgently required for achieving more efficient optical output from the OLETs, which possess the characteristics of a unique three-terminal device structure.

Balanced ambipolar charge transport is essential for the development of OLETs with high efficiency. In 2006, Sirringhaus and co-workers fabricated the first OLET in a bottom-contact/top-gate configuration using F8BT.[49] The achievement of a high EQE of 0.75% for F8BT-based OLET was attributed to a balanced ambipolar charge transport with hole and electron mobilities of 7.5×10^{-4} and 8.5×10^{-4} cm^2 V^{-1} s^{-1}, respectively, and a high PLQY (50–60% in solid films). Generally, high mobility emissive materials demonstrate typical hole or electron transport characteristics. An effective and universal strategy to achieve simultaneous injection of holes and electrons in unipolar materials is to construct an asymmetric geometry with different work function source and drain electrodes. The work function of the selected electrode materials should be as close as possible to the HOMO and LUMO levels of organic semiconductors to form a fine energy level matching for reducing the contact resistance, which is favorable for the formation of an ohmic contact. Thus, the holes and electrons can be more easily injected into the organic layers. As reported by Heeger et al., asymmetric electrodes can be successfully fabricated via angle evaporation technique.[111] The main purpose of this technique is to change the tilt angle of the substrate during the successive evaporation of two electrodes with different work functions. The asymmetric electrodes can also be prepared by changing the position of two different work function electrodes relative to the substrate in the evaporation process.[29] In these two examples, the fabrication of asymmetric electrodes is the fundamental reason for the successful fabrication of devices with balanced charge transportation based on p-type organic semiconductors. There are many other successful cases of fabrication of ambipolar OLETs based on asymmetric electrodes. Here, we have summarized some electrodes with different work functions for the injection of holes and electrons, which are commonly used in the OLETs, as shown in Figure 7d.[29,43,98,99] At present, there are only a few reported studies on *n*, active layer thickness, and gate materials. The optimization of these factors is essential to further improve the device performance, which has been discussed in Section 2.2

3.2. Current Advances in the OLET-Based Applications

AMOLED-based screens have been widely used in the preparation of smartphones. The self-luminescence characteristics of AMOLED make the devices thinner and less power consumption owing to the absence of backlight. AMOLED is constructed by controlling each light-emitting pixel with an independent thin-film transistor, which has a complex structure, difficult preparation process, and high cost. OLETs with unique device architectures that simultaneously integrate the switching functionalities of OFETs and the emission properties of OLEDs are expected to further simplify the device structure of AMOLEDs.

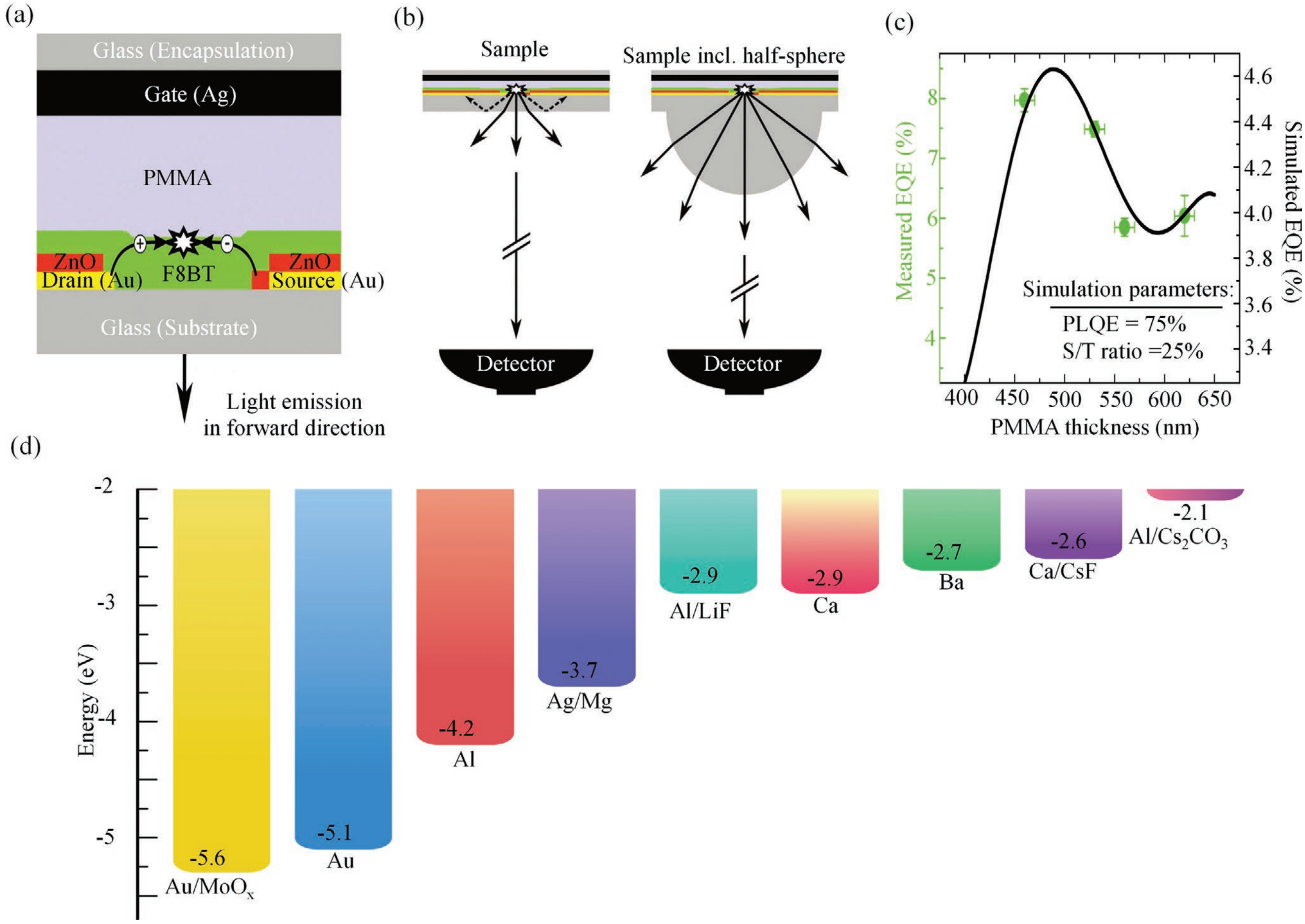

Figure 7. a) Schematic of bottom contact/top gate OLET with bottom-emitting based on F8BT. b) An illustration of the strategy for enhancing external light extraction out-coupling by adding half-sphere mounted in the direction of light-emitting. c) Measured and simulated EQE of OLET versus thickness of PMMA. d) Commonly used electrodes with different work functions for the injection of holes and electrons in OLETs. a–c) Reproduced with permission.[30] Copyright 2012, Wiley-VCH.

The aperture ratio, defined as the proportion of the light-emitting area of the device to the total area, is a major parameter of any display device. However, OLETs usually show a poor aperture ratio, especially ambipolar OLETs, due to the presence of electrodes and narrowed emission. In 2015, Namdas and coworkers presented hybrid area-emitting transistors with a satisfactory aperture rate of 50% (**Figure 8**a), which is far beyond the minimum required aperture ratio (34%) of AMOLED displays.[112] The active layer of the device consisted of an electron transport layer (zinc–tin oxide) and a light-emitting layer. Additionally, electron transport was dominant in the device. The achievement of an unprecedented aperture ratio with high brightness (Figure 8b) of this device was attributed to the unique nonplanar source/drain electrode with a transparent electrode on the top and circular design of the drain electrode. The characteristics of easy processing of organic semiconductors render them useful in fabricating OLETs with pixelated emission based on the patterned active layers. Recently, Park et al. demonstrated a red-emitting OLET based on Hex-4-TFPTA in pixelated emission.[62] This type of OLET is fabricated by the process of "patterned taping," a soft-lithographic technique, which exhibits a higher pixel resolution than that of a 4 K ultrahigh definition (UHD) 40 in. display screen (Figure 8c,d). However, even with the advances that have been made, the display application of OLETs is still a distant achievement because the simultaneous integration of high brightness, high efficiency, high aperture ratio, and low operating voltage in the devices remains a major challenge.

Organic semiconductor lasers are important devices in the field of organic optoelectronics. They have broad applications in the field of flexible wearable devices, intelligent interconnections, biomedicine, etc. Over the past years, optically pumped organic semiconductor lasers have received significant advances with tunable lasing colors, reduced laser thresholds, increased optical gains, etc.[11,25,113] However, optically pumped lasers usually require expensive equipment and a large pumping light source, which are unfavorable for the miniaturized device applications. The realization of electrically pumped organic lasers is of significant importance in science and technology because of their applications in miniaturized integrated optoelectronic circuits.[114] However, the realization of electrically pumped organic lasers remains a major scientific challenge because of the inevitable electrical and optical losses at high current densities and optical quenching induced by the electrical contacts. Over the past years, considerable efforts have been made in this field.[77,115,116] In comparison, the unique characteristics of

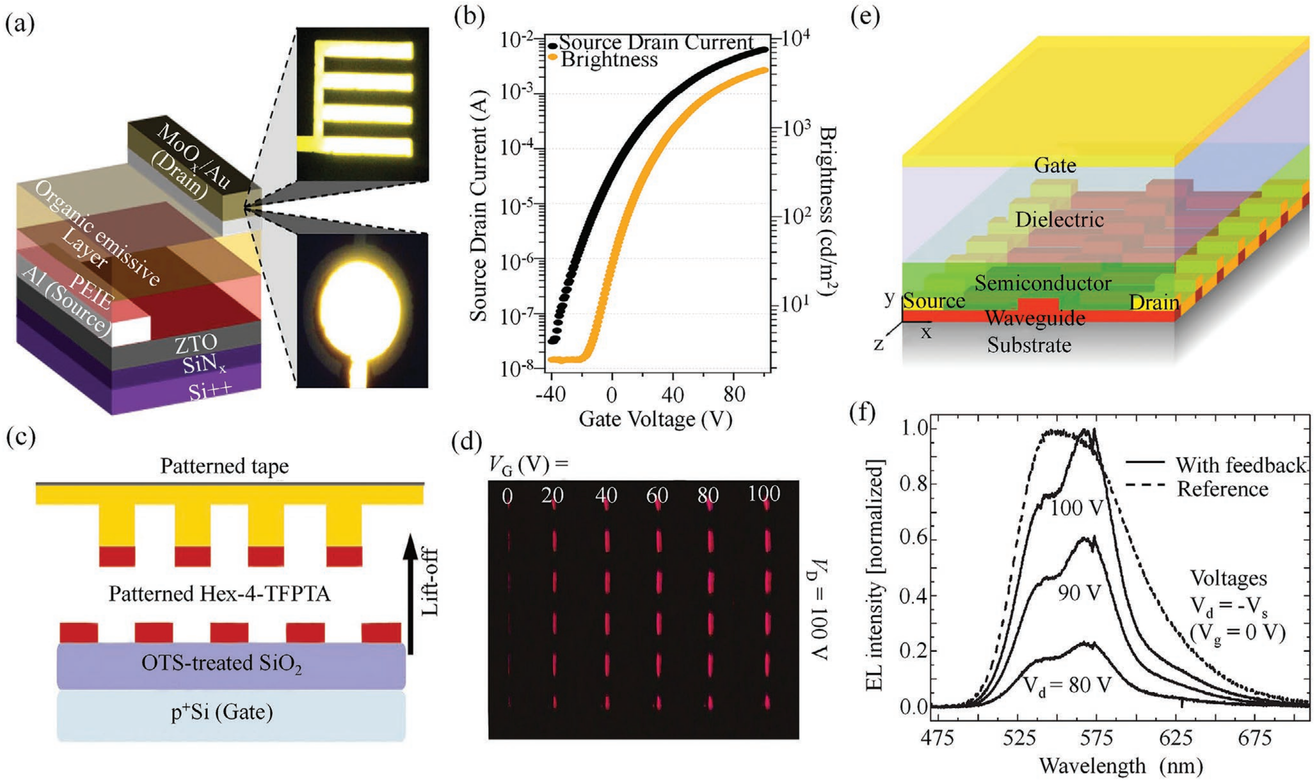

Figure 8. a) Schematic of OLET with area-emitting. b) The electrical and optical properties of the transistor as illustrated in (a) with a circular electrode. c) An illustration of the soft-lithographic technique. d) Electroluminescence images under different voltage conditions. ZTO = zinc tin oxide. e) Schematic illustration of the top-gate/bottom-contact device with integrated waveguide rib and distributed feedback grating. f) Electroluminescence spectra of the OLET for various drain/source voltages. a,b) Reproduced with permission.[112] Copyright 2015, Wiley-VCH. c,d) Reproduced with permission.[62] Copyright 2019, Wiley-VCH. e,f) Reproduced with permission.[115] Copyright 2009, Wiley-VCH.

spatially controllable light emission, far away from the electrodes, and high current density make OLETs promising candidates for the realization of electrically pumped organic semiconductor lasers since their emergence. Sirringhaus et al. fabricated a bottom-contact/top-gate OLET on a distributed feedback (DFB) substrate using Ta_2O_5 (Figure 8e).[115] The threshold values (4.5 µJ cm^{-2}) of the OLETs under optical pumping were comparable to those of the reference structures without metal electrodes, revealing that optical losses could be eliminated in this device. The authors concluded that only the narrowing of the electroluminescence spectrum (Figure 8f) was observed under the condition of *I–V*-sweep because it was about four orders of magnitude lower than the necessary singlet exciton density for the achievement of an electrically pumped laser in an OLET architecture. Recently, current-injection lasing based on organic semiconductors has been achieved in OLEDs with mixed-order DFB SiO_2 gratings.[77] Valuable fabrication guidelines have been obtained as a result of these efforts. However, for the realization of this important scientific goal, the development of superior high mobility organic semiconductor laser materials are crucial.[25,78,117]

OLETs, combining the characteristics of light emission and charge transport, have been considered as pivotal components for integrated circuits. However, the integration of multiple functions in a single device is still a major challenge. In recent years, significant developments have been made in device structure design and chemical structure engineering, because of which the integration of multiple functions in OLETs has attracted people's attention. In 2017, an organic up-conversion device was successfully fabricated by incorporating a near-infrared (NIR) absorption material between the charge transport layer and the light-emitting layer in a multilayer OLET.[32] The achievement of high up-conversion for this device is attributed to the gain mechanism, balanced photodetection, and efficient light emission of the OLET. Samorì and co-workers developed optically switchable OLETs (OSOLETs) by an ingenious combination of light-emitting polymer and photochromic molecules.[31] These photosensitive molecules present different states with different HOMO energy levels under irradiation with visible and ultra violet (UV) light (**Figure 9**a,c). Because of this changeable HOMO energy level, the transport of holes from light-emitting material to closed-form photochromic material was favored. However, this type of transportation did not occur in the open-form photochromic material. Therefore, Samorì et al. observed that when the device was irradiated by UV light, the photosensitive molecule was in the closed form and the device did not emit light. When the device was irradiated by visible light, the photosensitive molecule was in the open form and the device emitted light. Since light is the external control mode of OSOLETs, high-resolution light-emitting images can be created by patterned light, as shown in Figure 9d. These examples show the powerful ability of the OLETs to incorporate multiple functions, thereby allowing the development of new functional devices.

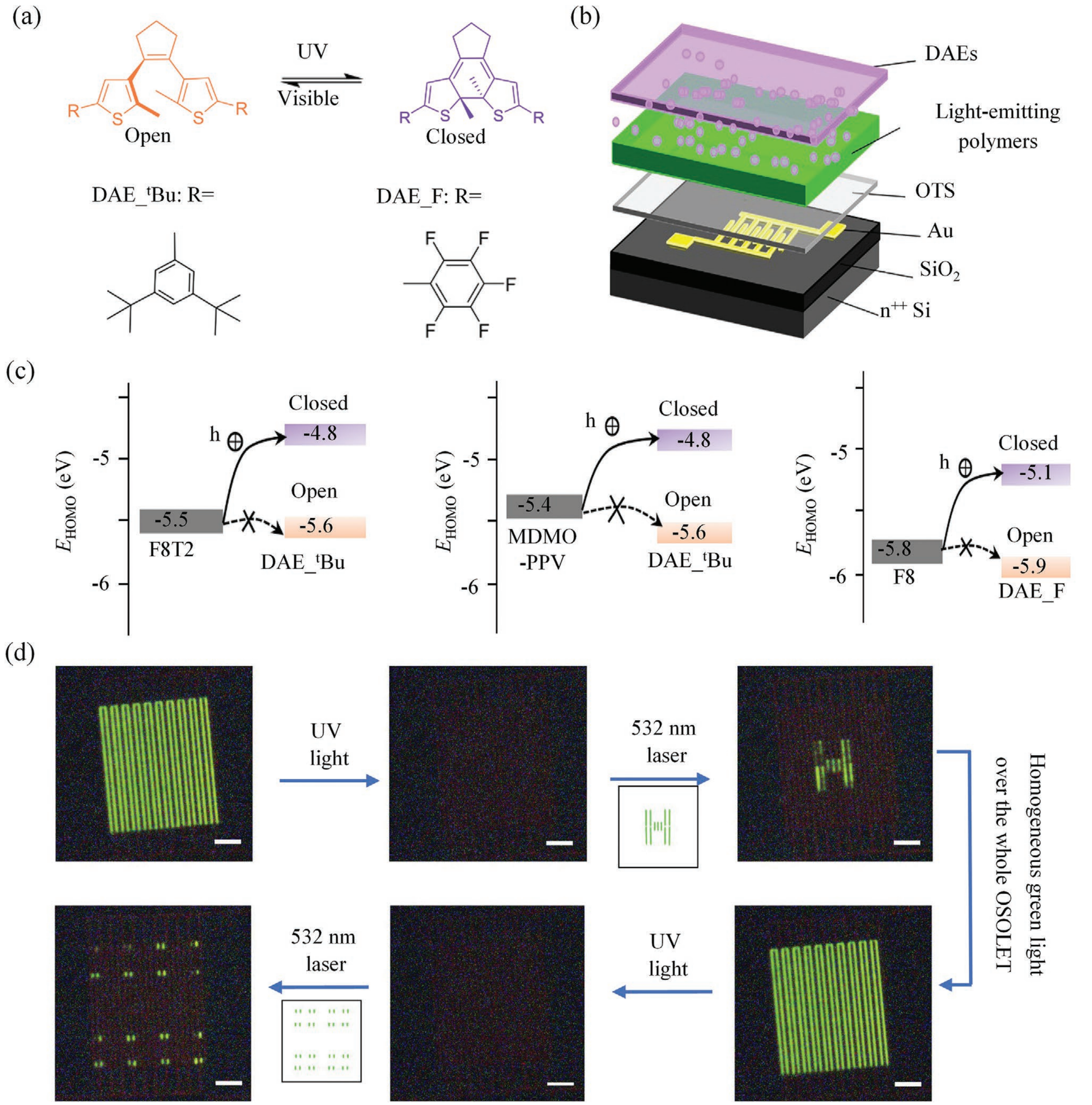

Figure 9. a) Chemical structures of photochromic molecules (DAE_tBu and DAE_F). b) Structure of the OSOLETs. c) Schematic illustration of the transport of holes from light-emitting material to photochromic material of open and closed form. d) High-resolution light-emitting images of OSOLET created by patterned light. The scale bar: 100 µm. a–d) Reproduced with permission.[31] Copyright 2019, Springer Nature.

4. Conclusions and Future Perspectives

In this review, we have provided a summary of the progress achieved in the field of OLETs with a special focus on the key points for developing high-efficiency OLETs from the aspect of materials and device optimization. In particular, the remarkable progress in OLETs along with the increased interest from researchers, may represent a new era of development in OLETs technology. From our point of view, to promote the rapid development of OLETs and their related fields, some challenges should be considered and overcome in the future: i) As mentioned before, high mobility emissive organic semiconductors are the ideal materials for OLETs because of their simplified and low-cost device fabrication, which aid in realizing the unique OLET characteristics. Some exciting progress has been recently made in this field; however, the efficiency of these materials still requires further improvements, including the charge transportation properties, PLQY, balanced transport of holes and electrons, and increased exciton utilization. Currently, the reported high mobility emissive organic semiconductors are mainly p-type dominating materials that demonstrate fluorescence emission properties. The development of n-type and even ambipolar high mobility organic semiconductors with high exciton utilization, such as phosphorescence (100%), TADF (≈100%), and TTA (62.5%) emission characteristics, is promising for further improvement in the OLET efficiency. ii) In addition, for multilayer OLETs, the selection of appropriate charge injection, charge transportation, and light-emitting layers with well-matched energies is crucial. Such a device structure is similar to that of the OLEDs and has good compatibility with the current OLED fabrication technology; however, the efficiency of the current OLETs is still much lower than the expected values with high driving voltages. In this case, the experience in the investigation of high-efficiency OLEDs would provide some valuable guidelines for the selection of ideal functional materials, device construction technology, optimizing interfacial quality between different layers, and improving the electroluminescent light transportation and output. In fact, only a few studies have been performed on the improvement of light transport and output from the aspect of device fabrication technology; thus, a considerable improvement is expected from this point. iii) To maintain a good gate field capacity with a high $I_{on/off}$ ratio and fast response, the insulator layer in the OLETs should also be modified, for instance, by incorporating high-κ dielectrics, such as SiNx and Al_2O_3 or by using a composite insulator layer, which is based on high-κ and low-κ insulators, for high charge carrier mobility. This is also favorable for low-voltage driving devices.[118,119] iv) The scientific goal of electrically pumped organic lasers, based on the OLETs, has not been definitely achieved until now, and extensive efforts should be made by scientists from different fields. In this case, developing high mobility low-threshold organic laser materials[25,61,117] and constructing appropriate optical resonators as well as developing novel device structures are crucial. v) Incorporating the characteristics of charge transport and light emission make the OLETs naturally advantageous in high-density logic applications and novel multifunctional photonic circuits.[31] However, this field of research has not received the required attention. vi) Compared to organic thin films, organic semiconductor single-crystals exhibit high charge carrier transportation and superior optical properties.[36,120] High-quality organic semiconductor single crystals, molecule-doped organic single crystals,[121] large-area arrays, and even large-area 2D crystals (including small molecules and conjugated polymers) with controllable thickness and heterojunction structures[122–127] would be promising candidates for the development of OLETs and their applications in integrated optoelectronic devices. The electrically driven organic photonic devices and circuits have the characteristics of high integration, light weight, and low cost.[10] To date, photonic devices and circuits based on OLETs have not been studied. As a device that can control both channel current and electroluminescence at the same time, it has inherent advantages when it is used as the driving source in photonic devices. Constructing photon paths on OLETs to realize efficient transmission and coupling between the photons and the devices is a challenging task. We believe that by overcoming the existing challenges in this field through the efforts of scientists from chemistry, material science, device physics, and organic photonics, a new rapid development stage for the OLETs will arrive, with significant improvements in the efficiency of the OLETs. These improved OLETs can be used for conducting fundamental studies and developing advanced display technology. Additionally, breakthroughs will be achieved in their applications in electrically pumped organic lasers and novel multifunctional integrated optoelectronic devices and circuits.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

display technology, electrical-driven organic photonic devices, electrically pumped organic lasers, high mobility, organic light-emitting transistors, strong emission

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[1] H. Dong, X. Fu, J. Liu, Z. Wang, W. Hu, *Adv. Mater.* **2013**, *25*, 6158.
[2] C. Wang, H. Dong, W. Hu, Y. Liu, D. Zhu, *Chem. Rev.* **2012**, *112*, 2208.
[3] H. Sirringhaus, *Adv. Mater.* **2014**, *26*, 1319.
[4] Y. Lin, Y. Li, X. Zhan, *Chem. Soc. Rev.* **2012**, *41*, 4245.
[5] Y. Cui, H. Yao, L. Hong, T. Zhang, Y. Tang, B. Lin, K. Xian, B. Gao, C. An, P. Bi, W. Ma, J. Hou, *Natl. Sci. Rev.* **2020**, *7*, 1239.
[6] X. Chen, X. Liu, M. A. Burgers, Y. Huang, G. C. Bazan, *Angew. Chem., Int. Ed.* **2014**, *53*, 14378.
[7] J.-X. Chen, W.-W. Tao, W.-C. Chen, Y.-F. Xiao, K. Wang, C. Cao, J. Yu, S. Li, F.-X. Geng, C. Adachi, C.-S. Lee, X.-H. Zhang, *Angew. Chem., Int. Ed.* **2019**, *58*, 14660.
[8] H. Uoyama, K. Goushi, K. Shizu, H. Nomura, C. Adachi, *Nature* **2012**, *492*, 234.
[9] Y. C. Liu, C. S. Li, Z. J. Ren, S. K. Yan, M. R. Bryce, *Nat. Rev. Mater.* **2018**, *3*, 18020.
[10] G. Zhao, H. Dong, Q. Liao, J. Jiang, Y. Luo, H. Fu, W. Hu, *Nat. Commun.* **2018**, *9*, 4790.
[11] Q. H. Cui, Q. Peng, Y. Luo, Y. Jiang, Y. Yan, C. Wei, Z. Shuai, C. Sun, J. Yao, Y. S. Zhao, *Sci. Adv.* **2018**, *4*, eaap9861.
[12] H. Wang, H. Liu, Q. Zhao, Z. Ni, Y. Zou, J. Yang, L. Wang, Y. Sun, Y. Guo, W. Hu, Y. Liu, *Adv. Mater.* **2017**, *29*, 1701772.
[13] Q. Zhang, *Front. Phys.* **2021**, *16*, 13602.
[14] A. Hepp, H. Heil, W. Weise, M. Ahles, R. Schmechel, H. von Seggern, *Phys. Rev. Lett.* **2003**, *91*, 157406.
[15] C. Zhang, P. Chen, W. Hu, *Small* **2016**, *12*, 1252.

[16] C. F. Liu, X. Liu, W. Y. Lai, W. Huang, *Adv. Mater.* **2018**, *30*, 1802466.
[17] M. U. Chaudhry, K. Muhieddine, R. Wawrzinek, J. Sobus, K. Tandy, S.-C. Lo, E. B. Namdas, *Adv. Funct. Mater.* **2020**, *30*, 1905282.
[18] D. Yuan, V. Sharapov, X. Liu, L. Yu, *ACS Omega* **2020**, *5*, 68.
[19] J. Zaumseil, *Adv. Funct. Mater.* **2020**, *30*, 1905269.
[20] M. Muccini, W. Koopman, S. Toffanin, *Laser Photonics Rev.* **2012**, *6*, 258.
[21] F. Cicoira, C. Santato, *Adv. Funct. Mater.* **2007**, *17*, 3421.
[22] J. Li, K. Zhou, J. Liu, Y. Zhen, L. Liu, J. Zhang, H. Dong, X. Zhang, L. Jiang, W. Hu, *J. Am. Chem. Soc.* **2017**, *139*, 17261.
[23] J. Liu, H. Zhang, H. Dong, L. Meng, L. Jiang, L. Jiang, Y. Wang, J. Yu, Y. Sun, W. Hu, A. J. Heeger, *Nat. Commun.* **2015**, *6*, 10032.
[24] Z. Xie, D. Liu, Y. Zhang, Q. Liu, H. Dong, W. Hu, *Chem. J. Chin. Univ.* **2020**, *41*, 1179.
[25] D. Liu, J. De, H. Gao, S. Ma, Q. Ou, S. Li, Z. Qin, H. Dong, Q. Liao, B. Xu, Q. Peng, Z. Shuai, W. Tian, H. Fu, X. Zhang, Y. Zhen, W. Hu, *J. Am. Chem. Soc.* **2020**, *142*, 6332.
[26] T. H. Thomas, D. J. Harkin, A. J. Gillett, V. Lemaur, M. Nikolka, A. Sadhanala, J. M. Richter, J. Armitage, H. Chen, I. McCulloch, S. M. Menke, Y. Olivier, D. Beljonne, H. Sirringhaus, *Nat. Commun.* **2019**, *10*, 2614.
[27] M. Chen, Y. Zhao, L. Yan, S. Yang, Y. Zhu, I. Murtaza, G. He, H. Meng, W. Huang, *Angew. Chem., Int. Ed.* **2017**, *56*, 722.
[28] J. Deng, Y. Xu, L. Liu, C. Feng, J. Tang, Y. Gao, Y. Wang, B. Yang, P. Lu, W. Yang, Y. Ma, *Chem. Commun.* **2016**, *52*, 2370.
[29] Z. Qin, H. Gao, J. Liu, K. Zhou, J. Li, Y. Dang, L. Huang, H. Deng, X. Zhang, H. Dong, W. Hu, *Adv. Mater.* **2019**, *31*, 1903175.
[30] M. C. Gwinner, D. Kabra, M. Roberts, T. J. Brenner, B. H. Wallikewitz, C. R. McNeill, R. H. Friend, H. Sirringhaus, *Adv. Mater.* **2012**, *24*, 2728.
[31] L. Hou, X. Zhang, G. F. Cotella, G. Carnicella, M. Herder, B. M. Schmidt, M. Patzel, S. Hecht, F. Cacialli, P. Samori, *Nat. Nanotechnol.* **2019**, *14*, 347.
[32] D. Li, Y. Hu, N. Zhang, Y. Lv, J. Lin, X. Guo, Y. Fan, J. Luo, X. Liu, *ACS Appl. Mater. Interfaces* **2017**, *9*, 36103.
[33] H. Chen, X. Xing, J. Miao, C. Zhao, M. Zhu, J. Bai, Y. He, H. Meng, *Adv. Opt. Mater.* **2020**, *8*, 1901651.
[34] H. Gao, J. Liu, Z. Qin, T. Wang, C. Gao, H. Dong, W. Hu, *Nanoscale* **2020**, *12*, 18371.
[35] S. Y. Cho, S. Y. Kim, S. Jeon, R. Choi, J. H. Lee, *Appl. Phys. Lett.* **2019**, *115*, 043301.
[36] X. Zhang, H. Dong, W. Hu, *Adv. Mater.* **2018**, *30*, 1801048.
[37] T. Takahashi, T. Takenobu, J. Takeya, Y. Iwasa, *Adv. Funct. Mater.* **2007**, *17*, 1623.
[38] L. Burgi, M. Turbiez, R. Pfeiffer, F. Bienewald, H. J. Kirner, C. Winnewisser, *Adv. Mater.* **2008**, *20*, 2217.
[39] F. Cicoira, C. Santato, A. Dadvand, C. Harnagea, A. Pignolet, P. Bellutti, Z. Xiang, F. Rosei, H. Meng, D. F. Perepichka, *J. Mater. Chem.* **2008**, *18*, 158.
[40] L. C. Ma, D. S. Qin, Y. Q. Liu, X. W. Zhan, *J. Mater. Chem. C* **2018**, *6*, 535.
[41] T. Takenobu, S. Z. Bisri, T. Takahashi, M. Yahiro, C. Adachi, Y. Iwasa, *Phys. Rev. Lett.* **2008**, *100*, 066601.
[42] T. Oyamada, H. Uchiuzou, H. Sasabe, C. Adachi, *J. Soc. Inf. Display* **2005**, *13*, 869.
[43] K. Tandy, M. Ullah, P. L. Burn, P. Meredith, E. B. Namdas, *Org. Electron.* **2013**, *14*, 2953.
[44] H. Nakanotani, R. Kabe, M. Yahiro, T. Takenobu, Y. Iwasa, C. Adachi, *Appl. Phys. Express* **2008**, *1*, 091801.
[45] J. Zaumseil, R. H. Friend, H. Sirringhaus, *Nat. Mater.* **2006**, *5*, 69.
[46] T. H. Ke, R. Gehlhaar, C. H. Chen, J. T. Lin, C. C. Wu, C. Adachi, *Appl. Phys. Lett.* **2009**, *95*, 063303.
[47] T. Sakanoue, M. Yahiro, C. Adachi, H. Uchiuzou, T. Takahashi, A. Toshimitsu, *Appl. Phys. Lett.* **2007**, *90*, 171118.
[48] J. Zaumseil, C. R. McNeill, M. Bird, D. L. Smith, P. P. Ruden, M. Roberts, M. J. McKiernan, R. H. Friend, H. Sirringhaus, *J. Appl. Phys.* **2008**, *103*, 064517.
[49] J. Zaumseil, C. L. Donley, J. S. Kim, R. H. Friend, H. Sirringhaus, *Adv. Mater.* **2006**, *18*, 2708.
[50] T. Oyamada, C. H. Chang, T. C. Chao, F. C. Fang, C. C. Wu, K. T. Wong, H. Sasabe, C. Adachi, *J. Phys. Chem. C* **2007**, *111*, 108.
[51] T. Kono, D. Kumaki, J. Nishida, T. Sakanoue, M. Kakita, H. Tada, S. Tokito, Y. Yamashita, *Chem. Mater.* **2007**, *19*, 1218.
[52] S. Z. Bisri, T. Takenobu, Y. Yomogida, H. Shimotani, T. Yamao, S. Hotta, Y. Iwasa, *Adv. Funct. Mater.* **2009**, *19*, 1728.
[53] H. Nakanotani, M. Saito, H. Nakamura, C. Adachi, *Appl. Phys. Lett.* **2009**, *95*, 103307.
[54] A. Dadvand, A. G. Moiseev, K. Sawabe, W. H. Sun, B. Djukic, I. Chung, T. Takenobu, F. Rosei, D. F. Perepichka, *Angew. Chem., Int. Ed.* **2012**, *51*, 3837.
[55] K. Niimi, H. Mori, E. Miyazaki, I. Osaka, H. Kakizoe, K. Takimiya, C. Adachi, *Chem. Commun.* **2012**, *48*, 5892.
[56] Y. Yomogida, T. Takenobu, H. Shimotani, K. Sawabe, S. Z. Bisri, T. Yamao, S. Hotta, Y. Iwasa, *Appl. Phys. Lett.* **2010**, *97*, 173301.
[57] K. Oniwa, T. Kanagasekaran, T. N. Jin, M. Akhtaruzzaman, Y. Yamamoto, H. Tamura, I. Hamada, H. Shimotani, N. Asao, S. Ikeda, K. Tanigaki, *J. Mater. Chem. C* **2013**, *1*, 4163.
[58] Y. Yomogida, H. Sakai, K. Sawabe, S. Gocho, S. Z. Bisri, H. Nakanotani, C. Adachi, T. Hasobe, Y. Iwasa, T. Takenobu, *Org. Electron.* **2013**, *14*, 2737.
[59] T. Komori, H. Nakanotani, T. Yasuda, C. Adachi, *J. Mater. Chem. C* **2014**, *2*, 4918.
[60] M. Zambianchi, E. Benvenuti, C. Bettini, C. Zanardi, R. Seeber, D. Gentili, M. Cavallini, M. Muccini, V. Biondo, C. Soldano, G. Generali, S. Toffanin, M. Melucci, *J. Mater. Chem. C* **2016**, *4*, 9411.
[61] S. Q. Ma, K. Zhou, M. X. Hu, Q. Y. Li, Y. J. Liu, H. T. Zhang, J. B. Jing, H. L. Dong, B. Xu, W. P. Hu, W. J. Tian, *Adv. Funct. Mater.* **2018**, *28*, 1802454.
[62] S. Oh, J. H. Kim, S. K. Park, C. H. Ryoo, S. Y. Park, *Adv. Opt. Mater.* **2019**, *7*, 1901274.
[63] A. Dadvand, W. H. Sun, A. G. Moiseev, F. Belanger-Gariepy, F. Rosei, H. Meng, D. F. Perepichka, *J. Mater. Chem. C* **2013**, *1*, 2817.
[64] M. Melucci, L. Favaretto, M. Zambianchi, M. Durso, M. Gazzano, A. Zanelli, M. Monari, M. G. Lobello, F. De Angelis, V. Biondo, G. Generali, S. Troisi, W. Koopman, S. Toffanin, R. Capelli, M. Muccini, *Chem. Mater.* **2013**, *25*, 668.
[65] L. Zheng, J. F. Li, K. Zhou, X. X. Yu, X. T. Zhang, H. L. Dong, W. P. Hu, *Nano Res.* **2020**, *13*, 1976.
[66] D. Liu, C. G. Li, S. J. Niu, Y. Li, M. X. Hu, Q. Y. Li, W. G. Zhu, X. T. Zhang, H. L. Dong, W. P. Hu, *J. Mater. Chem. C* **2019**, *7*, 5925.
[67] J. Li, J. Liu, Y. G. Zhen, L. Q. Meng, Y. Wang, H. L. Dong, W. P. Hu, *J. Mater. Chem. C* **2015**, *3*, 10695.
[68] J. F. Li, L. Zheng, L. J. Sun, C. G. Li, X. T. Zhang, S. S. Cheng, W. P. Hu, *J. Mater. Chem. C* **2018**, *6*, 13257.
[69] B. K. Yap, R. Xia, M. Campoy-Quiles, P. N. Stavrinou, D. D. C. Bradley, *Nat. Mater.* **2008**, *7*, 376.
[70] Z. Zhao, S. M. Gao, X. Y. Zheng, P. F. Zhang, W. T. Wu, R. T. K. Kwok, Y. Xiong, N. L. C. Leung, Y. C. Chen, X. K. Gao, J. W. Y. Lam, B. Z. Tang, *Adv. Funct. Mater.* **2018**, *28*, 1705609.
[71] Q. Liu, Y. Zhang, C. Gao, T. Wang, W. Hu, H. Dong, *Acta Chim. Sinica* **2020**, *78*, 945.
[72] Y. Zhang, J. Ye, Z. Liu, Q. Liu, X. Guo, Y. Dang, J. Zhang, Z. Wei, Z. Wang, Z. Wang, H. Dong, W. Hu, *J. Mater. Chem. C* **2020**, *8*, 10868.
[73] H. Zhang, Z. Zhao, A. T. Turley, L. Wang, P. R. McGonigal, Y. Tu, Y. Li, Z. Wang, R. T. K. Kwok, J. W. Y. Lam, B. Z. Tang, *Adv. Mater.* **2020**, *32*, 2001457.

[74] J. Cornil, D. A. dos Santos, X. Crispin, R. Silbey, J. L. Bredas, *J. Am. Chem. Soc.* **1998**, *120*, 1289.
[75] J. Cornil, D. Beljonne, J. P. Calbert, J. L. Bredas, *Adv. Mater.* **2001**, *13*, 1053.
[76] C. Gao, S. K. K. Prasad, B. L. Zhang, M. Dvorak, M. J. Y. Tayebjee, D. R. McCamey, T. W. Schmidt, T. A. Smith, W. W. H. Wong, *J. Phys. Chem. C* **2019**, *123*, 20181.
[77] A. S. D. Sandanayaka, T. Matsushima, F. Bencheikh, S. Terakawa, W. J. Potscavage, C. J. Qin, T. Fujihara, K. Goushi, J. C. Ribierre, C. Adachi, *Appl. Phys. Express* **2019**, *12*, 061010.
[78] Q. Ou, Q. Peng, Z. Shuai, *Nat. Commun.* **2020**, *11*, 4485.
[79] C. Rost, S. Karg, W. Riess, M. A. Loi, M. Murgia, M. Muccini, *Appl. Phys. Lett.* **2004**, *85*, 1613.
[80] F. Dinelli, R. Capelli, M. A. Loi, M. Murgia, M. Muccini, A. Facchetti, T. J. Marks, *Adv. Mater.* **2006**, *18*, 1416.
[81] M. Ullah, K. Tandy, S. D. Yambem, M. Aljada, P. L. Burn, P. Meredith, E. B. Namdas, *Adv. Mater.* **2013**, *25*, 6213.
[82] D. K. Kim, J. D. Oh, E. S. Shin, H. S. Seo, J. H. Choi, *J. Appl. Phys.* **2014**, *115*, 164503.
[83] J. Sobus, F. Bencheikh, M. Mamada, R. Wawrzinek, J. C. Ribierre, C. Adachi, S. C. Lo, E. B. Namdas, *Adv. Funct. Mater.* **2018**, *28*, 1800340.
[84] M. Ullah, R. Wawrzinek, F. Maasoumi, S. C. Lo, E. B. Namdas, *Adv. Opt. Mater.* **2016**, *4*, 1022.
[85] Y. S. Hu, L. Song, D. W. Li, J. Lin, X. Y. Lui, *Org. Electron.* **2015**, *26*, 92.
[86] M. Prosa, E. Benvenuti, M. Pasini, U. Giovanella, M. Bolognesi, L. Meazza, F. Galeotti, M. Muccini, S. Toffanin, *ACS Appl. Mater. Interfaces* **2018**, *10*, 25580.
[87] M. Ullah, K. Tandy, A. J. Clulow, P. L. Burn, I. R. Gentle, P. Meredith, S. C. Lo, E. B. Namdas, *ACS Photonics* **2017**, *4*, 754.
[88] J. H. Seo, E. B. Namdas, A. Gutacker, A. J. Heeger, G. C. Bazan, *Adv. Funct. Mater.* **2011**, *21*, 3667.
[89] D. W. Li, Y. S. Hu, X. Y. Guo, Y. Lv, N. Zhang, J. Lin, X. Y. Liu, *J. Phys. Chem. C* **2018**, *122*, 15190.
[90] D. K. Kim, Y. L. Kim, J. H. Choi, *J. Phys. Chem. C* **2019**, *123*, 11063.
[91] C. Soldano, R. D'Alpaos, G. Generali, *ACS Photonics* **2017**, *4*, 800.
[92] J. H. Lee, T. H. Ke, J. Genoe, P. Heremans, C. Rolin, *Adv. Electron. Mater.* **2019**, *5*, 1800437.
[93] R. Capelli, S. Toffanin, G. Generali, H. Usta, A. Facchetti, M. Muccini, *Nat. Mater.* **2010**, *9*, 496.
[94] H. S. Seo, Y. Zhang, M. J. An, J. H. Choi, *Org. Electron.* **2009**, *10*, 1293.
[95] C. Soldano, G. Generali, E. Cianci, G. Tallarida, M. Fanciulli, M. Muccini, *Thin Solid Films* **2016**, *616*, 408.
[96] R. Wawrzinek, K. Muhieddine, M. Ullah, P. B. Koszo, P. E. Shaw, A. Grosjean, F. Maasoumi, D. M. Stoltzfus, J. K. Clegg, P. L. Burn, E. B. Namdas, S. C. Lo, *Adv. Opt. Mater.* **2016**, *4*, 1867.
[97] M. A. McCarthy, B. Liu, E. P. Donoghue, I. Kravchenko, D. Y. Kim, F. So, A. G. Rinzler, *Science* **2011**, *332*, 570.
[98] K. Obara, S. Higashihara, T. Yamao, S. Hotta, *Jpn. J. Appl. Phys.* **2016**, *55*, 03DC09.
[99] D. K. Kim, J. D. Oh, J. W. Kim, H. Y. Lee, J. H. Choi, *J. Phys. Chem. C* **2016**, *120*, 13716.
[100] B. B. Hsu, C. Duan, E. B. Namdas, A. Gutacker, J. D. Yuen, F. Huang, Y. Cao, G. C. Bazan, I. D. Samuel, A. J. Heeger, *Adv. Mater.* **2012**, *24*, 1171.
[101] C. Adachi, A. S. D. Sandanayaka, *CCS Chem.* **2020**, *2*, 1203.
[102] S. Z. Bisri, K. Sawabe, M. Imakawa, K. Maruyama, T. Yamao, S. Hotta, Y. Iwasa, T. Takenobu, *Sci. Rep.* **2012**, *2*, 985.
[103] K. Yamada, T. Yamao, S. Hotta, *Adv. Mater.* **2013**, *25*, 2860.
[104] K. Sawabe, M. Imakawa, M. Nakano, T. Yamao, S. Hotta, Y. Iwasa, T. Takenobu, *Adv. Mater.* **2012**, *24*, 6141.
[105] K. Zhou, J. Liu, H. Dong, S. Ding, Y. Zhen, Y. Shi, W. Hu, *J. Mater. Chem. C* **2020**, *8*, 16333.
[106] Y. Wan, J. Deng, W. Wu, J. Zhou, Q. Niu, H. Li, H. Yu, C. Gu, Y. Ma, *ACS Appl. Mater. Interfaces* **2020**, *12*, 43976.
[107] E. B. Namdas, B. B. Y. Hsu, Z. Liu, S. C. Lo, P. L. Burn, I. D. W. Samuel, *Adv. Mater.* **2009**, *21*, 4957.
[108] L. Song, Y. Hu, Z. Liu, Y. Lv, X. Guo, X. Liu, *ACS Appl. Mater. Interfaces* **2017**, *9*, 2711.
[109] A. Kim, G. Huseynova, J. Lee, J. H. Lee, *Org. Electron.* **2019**, *71*, 246.
[110] E. B. Namdas, B. B. Hsu, J. D. Yuen, I. D. Samuel, A. J. Heeger, *Adv. Mater.* **2011**, *23*, 2353.
[111] J. S. Swensen, C. Soci, A. J. Heeger, *Appl. Phys. Lett.* **2005**, *87*, 253511.
[112] K. Muhieddine, M. Ullah, F. Maasoumi, P. L. Burn, E. B. Namdas, *Adv. Mater.* **2015**, *27*, 6677.
[113] I. D. Samuel, G. A. Turnbull, *Chem. Rev.* **2007**, *107*, 1272.
[114] A. J. Heeger, N. S. Sariciftci, E. B. Namdas, *Semiconducting and Metallic Polymers*, Oxford University Press, New York **2010**.
[115] M. C. Gwinner, S. Khodabakhsh, M. H. Song, H. Schweizer, H. Giessen, H. Sirringhaus, *Adv. Funct. Mater.* **2009**, *19*, 1360.
[116] K. Maruyama, K. Sawabe, T. Sakanoue, J. Li, W. Takahashi, S. Hotta, Y. Iwasa, T. Takenobu, *Sci. Rep.* **2015**, *5*, 10221.
[117] M. Ichikawa, K. Nakamura, M. Inoue, H. Mishima, T. Haritani, R. Hibino, T. Koyama, Y. Taniguchi, *Appl. Phys. Lett.* **2005**, *87*, 221113.
[118] F. Yang, L. Sun, J. Han, B. Li, X. Yu, X. Zhang, X. Ren, W. Hu, *ACS Appl. Mater. Interfaces* **2018**, *10*, 25871.
[119] D. K. Kim, J. H. Choi, *Org. Electron.* **2017**, *51*, 287.
[120] Y. Wang, J. Yang, Y. Gong, M. Fang, Z. Li, B. Z. Tang, *SmartMat* **2020**, *1*, e1006.
[121] Z. Qin, C. Gao, W. W. H. Wong, M. K. Riede, T. Wang, H. Dong, Y. Zhen, W. Hu, *J. Mater. Chem. C* **2020**, *8*, 14996.
[122] J. Wu, Q. Li, G. Xue, H. Chen, H. Li, *Adv. Mater.* **2017**, *29*, 1606101.
[123] Y. Shi, L. Jiang, J. Liu, Z. Tu, Y. Hu, Q. Wu, Y. Yi, E. Gann, C. R. McNeill, H. Li, W. Hu, D. Zhu, H. Sirringhaus, *Nat. Commun.* **2018**, *9*, 2933.
[124] C. Xu, P. He, J. Liu, A. Cui, H. Dong, Y. Zhen, W. Chen, W. Hu, *Angew. Chem., Int. Ed.* **2016**, *55*, 9519.
[125] Y. Yao, H. Dong, F. Liu, T. P. Russell, W. Hu, *Adv. Mater.* **2017**, *29*, 1701251.
[126] Y. Yao, Y. Chen, H. Wang, P. Samorì, *SmartMat* **2020**, *1*, e1009.
[127] H. Dong, W. Hu, *Acc. Chem. Res.* **2016**, *49*, 2435.

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