Organic Light-Emitting Transistors: Advances and Perspectives

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Abstract

The rapid development of charge transporting and light-emitting materials in the last decades has advanced device performance, highlighting the high potential of light-emitting transistors (LETs). Demonstrated for the first time over 15 years ago, LETs have transformed from an optoelectronic curiosity to a serious competitor in the race for cheaper and more efficient displays, also showing promise for injection lasers. So, what is an LET, how does it work and what are the current challenges for its integration into mainstream technologies? This progress report aims to shed some light on these questions. This work will also be looking at the fundamental working principle of LETs, materials that have been used, and device physics and architectures involved in the progression of LET technology. State-of-the-art development for LETs will be explored as prospect avenues for the future of research and applications in this area.

1. Introduction

Like many technologies, the exact origins of light-emitting transistors (LETs) are difficult to be clearly defined. Among the earliest reported work, a study carried out at the Interuniversity Microelectronics Center in Leuven, Belgium[1] reported an InGaAs bipolar resonant tunnelling transistor, enabling light emission at liquid nitrogen temperatures. That report cited an even earlier study by L. Eaves,[2] regarding resonant tunnelling phenomena. As it stands, early efforts have mainly revolved around suppressing optical emissive properties of the devices in order to enhance their transistor characteristics.

Two studies, however, stand out as milestones in this field. The first is a demonstration of light emission from a three-terminal bipolar heterojunction transistor by Holonyak *et al.*[3] The device was fabricated from indium gallium phosphide, indium gallium arsenide, and gallium arsenide to produce infrared photons.[3] The second milestone study published just a few months earlier by Hepp *et al.*[4] set the stage for the 15 years of research to come by presenting the first organic LET based on an evaporated tetracene layer with a simple device structure. This device distinguished itself not just because of the active organic semiconductor layer, but also because light emission was well inside the visible spectrum. This study was the launch pad for a much bigger race that would follow in pursuing different LET architectures and applying them to display technology as well as possibly injection lasers. This review is mainly focused on the progress of organic-based LETs, first explaining how an LET works, the device physics and architectures involved, what materials have been explored and what are the current main challenges as well as some new prospects for possibly advancement and integration into mainstream technologies.

So, what is a light-emitting transistor? An LET combines the switching abilities of a transistor with the emissive properties of an LED. The result is a multifunctional device of either unipolar (electron or hole majority carriers) or ambipolar nature (electron and hole majority carriers) with an optical emission. Such dual functionality places LETs in a favourable position for several applications. Firstly, LETs provide a unique opportunity to explore the charge transport, charge injection and optical characteristics of device architectures and materials simultaneously. This is highly relevant for a rapidly changing field of applied physics today that frequently intersects with material science and the engineering, and testing of new chromophores for such applications. Secondly, LETs are positioned well to enter the rich domain of integrated circuitry and high-speed signal processing that involves both electrical signals and optical signals. While a standard transistor has an electrical input and output function only, an LET processes a third component in the form of optical output. The additional functionality gives LETs the prospective to revolutionise the way that communications circuitry is built. Thirdly, LETs could potentially be the avenue through which electrically pumped lasing is finally achieved. This is mainly because of their ability to provide high current densities and low optical losses at the electrodes. Finally, and perhaps the most popularly cited application of LETs, is their easy integration into display circuitry. With the intrinsic combination of a transistor and an organic light-emitting diode (OLED) and the vast quantity of available and rapidly changing materials available, LETs could be used to simplify the circuitry required for display applications. With the success in employing metal oxides in certain LET architectures, this could potentially eliminate the need of the costly processing silicon that is usually used as the backplane for light-emitting technology.

2. LET Characteristics and Working Mechanism

Just as it combines the dual-functionality of a transistor and an OLED, so is an LET characterised in the same fashion as both of these two types of devices. The charge transport characteristics are determined using the same combination of transfer and output characteristics that are used in a standard transistor. Key characteristics such as device mobility, threshold voltage, contact resistance, operating voltages and ON/OFF ratio are vital to ensure that the measured LET is living up to its switching capabilities.

Optically, similar characteristics as those of an OLED are extracted and calculated, namely the external quantum efficiency (EQE) of the device, and the brightness. The EQE is the ratio of the emitted photons, reaching the viewer from the front of the device, out of the device to charge carriers injected into the device. Ideally, devices will have both a high EQE and high brightness. This is a challenge in a LET geometry since semiconductor materials with a high photoluminescence quantum yield (PLQY) often have a low field-effect mobility (*i.e.*, charge carrier mobility). Conversely, many high mobility materials have suffered from a low PLQY due to strong intermolecular interaction. A high mobility can help improve the device brightness since charges move through the organic semiconductor faster to recombine. This in general from material point of view is a trade-off.

Of significant importance is the evolution of the electrical and optical characteristics with the applied gate and source-drain voltage in the device. Depending upon the relative electron and hole mobilities of the active organic semiconductor material and the choice of device architecture, unipolar or ambipolar LETs can be created. The device operation of both unipolar and ambipolar devices is described in the following sections.

2.1 Unipolar LETs

For unipolar LETs, charge transport within the device is either hole- or electron-dominated. Only one type of charge carrier is transported within the channel even though both electrons and holes must be injected for recombination and light emission. For a hole-dominated device, holes are injected into the device and are transported through the transistor channel. Electrons are injected, but remain close to the electrode since electron transport through the semiconductor channel is poor in such hole-dominated devices. Consequently, recombination and emission will, therefore, occur close to the electron-injecting electrode [*i.e.*, electrode with low work-function (LWF) see **Figure 1**]. The emission zone will be stationary during device operation since the charge transport is always dominated by holes. Operation in an electron-dominated device is the same except the polarity of the gate voltage is positive instead of negative, electrons are accumulated within the channel, and charge recombination and emission occur close to the hole-injecting electrode [*i.e.*, electrode with high workfunction (HWF) see **Figure 1**]. The first reported organic LET was a tetracene-based unipolar device and operated in a hole-accumulation mode.[4] While initially, unipolar organic LETs commonly used symmetric source-drain electrodes,[5-7] the popularity of asymmetric source-drain electrodes with metals of differing work functions increased rapidly. These lowered the barrier of both electrons and holes injection into LETs, increasing device efficiency.[8-10]

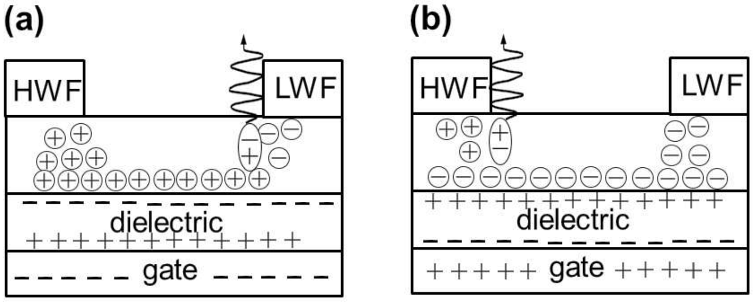

Figure 1: Unipolar LET operation in **a)** hole accumulation mode. In this mode, holes and electrons are both injected into the device from the respective source and drain contacts but only holes may accumulate within the channel. Light emission occurs next to the electron-injecting electrode with a low work function; **b)** electron accumulation mode. In this mode, both types of charges are injected into the device but only electrons accumulate within the channel and light emission occurs next to the electron-injecting electrode with a high work function.

2.2 Ambipolar LETs

Ambipolar LETs were also reported using a single-layer organic semiconductor.[11-13] In all three of these reports, the emission zone could be shifted through the channel using different applied gate voltages. An EQE of ≈0.75% was obtained using poly(9,9-di-*n*-octyl-fluorene-*alt*-

benzothiadiazole (F8BT) as the active layer.[13] For ambipolar LETs, both electrons and holes can accumulate in the channel, depending on the polarity of the applied voltage [14]. Ambipolar LETs can be achieved by using devices with asymmetric source-drain electrodes with a high work function and low work function electrode (see **Figure 2**). As with unipolar devices, electron and hole injection are also possible using a high work function electrode only in some cases. An example of the electrical and optical transfer characteristics for an ambipolar LET can be found in **Figure 2**. There are three distinct electron-dominated and hole-dominated regimes, with a cross-over regime, often called the 'ambipolar regime':

i) *Large positive gate voltage (V_G)*

When the V_G is large and positive, the charge transport is dominated by electrons and electrons are dominated within the transistor channel (see **Figure 2a**). Holes accumulate near the hole-injecting contact. Electrons may recombine with holes in the vicinity of the hole-injecting contact to emit light.[11] The brightness of the device is at a maximum since the current is high. The charge carriers are not balanced in this region, and the ratio of charges injected into the device for recombination and light emission is low. This results in a low EQE.[15]

ii) *Small V_G (close to zero)*

As the gate voltage becomes closer to zero, the electron accumulation within the channel decreases, and the electron density no longer extends across the whole channel. At the same time, since the gate voltage is less positive, and moving in the negative direction, holes are better able to accumulate, and the density of holes begins to build up and extend further into the channel. The electron and hole currents begin to become more balanced. When hole currents and electron currents are exactly balanced, light emission will occur in the middle of the transistor channel (see **Figure 2c**). The brightness here is low since the current in this region is also low. However, the number of electrons and holes is more balanced within the device and hence the EQE is increased.[15]

iii) *Large negative V_G*

When the gate voltage is at a large negative bias, mainly holes accumulate within the channel, with the density extending across the transistor channel towards the electron-injecting contact. Electrons accumulate mainly close to the low work function contact (see **Figure 2d**). The emission zone will be close to the electron injecting contact.[16] The EQE is again low since the charge carriers become less balanced within the channel.

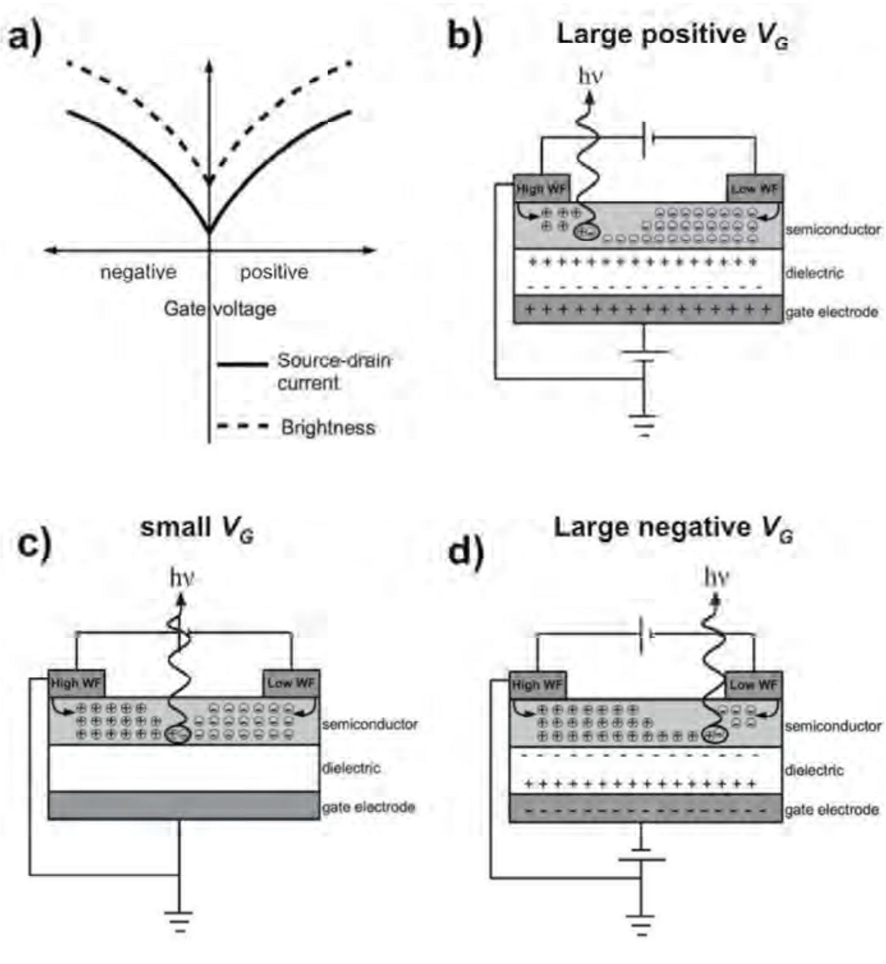

Figure 2. An example of ambipolar LET device characteristics operation. **a)** Typical electrical and optical transfer characteristics for an ambipolar LET; **b)** the application of a large positive V_G causes mainly electrons to accumulate within the transistor channel and the light emission occurs close to the high work function, hole-injecting electrode; **c)** the application of a small V_G increases the hole accumulation and so the charges recombine closer to the middle of the transistor channel and away from the electrodes; **d)** application of a large negative V_G causes mainly holes to accumulate in the transistor channel and light emission occurs close to the low work function, electron-injecting electrode.

3. Single Layer LETs

Thus far the LETs discussed have been constructed with a single, organic semiconductor layer that served the dual purpose of charge carrier and emissive layer. These devices can be viewed as the first generation of LETs, and were made use of a variety of material classes such as solution processed polymers, evaporated small organic molecules and single crystals (see **Table 1**). A limiting factor of these first generation LETs was the tendency towards a trade-off between the electrical and the optical characteristics, namely the mobility and the EQE. For example, an evaporated small organic molecule such as tetracene offers a higher mobility, but that advantage is countered by a low optical output.[4] This is of course due to the nature of the optoelectronic interactions in the device, namely low PLQY, excitonic quenching due to close proximity to higher charge carrier densities, and the limitation that higher charge carrier mobilities and shorter transit time put on the recombination rate in the LETs.

Table 1. Summary of the various single layer LETs and their device performance

Single layer LETs							
Ref. #	**Material**	**Color**	μ_{hole} ($cm^2 V^{-1} s^{-1}$)	$\mu_{electron}$ ($cm^2 V^{-1} s^{-1}$)	**ON/OFF Ratio**	**Max Power** (W) / **max Luminance** ($cd\ m^{-2}$)	**EQE** (%) or cd/A **at max Luminance**
[13]	F8BT	Green-Yellow	0.0006	0.00005	<100	–	0.6
[17]	F8BT (split-gate)	Green-Yellow	0.000094	0.000025	<100	609 $cd\ m^{-2}$	1.3
[18]	F8BT	Green	<0.001	<0.001	<10	8,000 $cd\ m^{-2}$	4
[19]	rubrene & tetracene (single crystals)	Orange & Green	0.82 & 2.3	0.27 & 0.12	$\approx 10^2$ & $\approx 10^5$		0.002 & 0.02
[20]	EFIN	Blue	0.000006	–	$\approx 10^4$	–	0.2
[21]	BSB-Me (single crystal)	Blue	0.0001	0.01	$\approx 10^3$	–	0.2
[22]	P3V2 (single crystal)	Blue	0.11	0.013	$\approx 10^4$	–	0.1
[23]	PDY-132	Yellow	≈0.0003	≈0.0003	$\approx 10^4$	43 $cd\ m^{-2}$	0.19
[24]	F8BT	Green-Yellow	0.0007–0.0009	0.0007–0.0009	<100	–	0. 75
[25]	F8TBT	Red	0.0005	0.00003	<100	80 nW	0. 4
[6]	MEH-PPV	Yellow-Red	–	–	10^5	–	–
[4]	Tetracene (film)	Yellow	0.005	–	10^6	45 $cd\ m^{-2}$	0.0024 cd/A
[26]	BTBT-C_{10}	UV	6	–	$\approx 10^9$	≈1 μW	0.003
	Ph-BTBT-C_{10}	Blue	2	–	$\approx 10^7$	≈0.08 μW	0.0006

4. Multilayer LETs

To overcome the trade-off problem of electrical and the optical characteristics in a single layer device with just one active organic material, a second generation of LETs (**Figure 3**) comprised of bilayer or multiple organic semiconductor films were developed. Multilayer architectures allowed the assignment of the charge transport and emission responsibilities to separate layers so that the electrical and optical properties of the resulting LETs could be addressed individually. **Figure 3** shows the bi-layer and tri-layer device architecture. For example, an emissive polymer Super Yellow was used in combination with a charge transport layer of poly(2,5-bis(3-alkylthiophene-2-yl)thieno[3,2-b]thiophene) (PBTTT) to obtain a maximum brightness of 2,500 cd m^{-2}.[27] The maximum EQE of 0.15% of the Super Yellow/PBTTT bilayer devices [27] was lower than the maximum EQE obtained for single layer Super Yellow devices.[10] The bilayer devices operated in p-type mode only, whereas the single layer devices were ambipolar. The lower EQE in the bilayer devices was due to the imbalance of charge carriers within the device.

A conjugated polyelectrolyte[29] and Cs_2CO_3[30] layer was used to improve the electron injection into bilayer Super Yellow/PBTTT devices. The conjugated polyelectrolyte allowed high work function electrodes to be used instead of the asymmetric electrodes used in other reports. This simplified device fabrication over previously reported devices since low work function metals were not needed.[27] Further improvements to this device architecture could be made using a more ordered polymer charge transport layer with a higher mobility than PBTTT as the charge transporting polymer.[31] In this case, the gate dielectric was structured using directional rubbing. This resulted in an increase in the mobility of the charge transporting polymer, poly(3,6-di-alkylthieno[3,2-b]thiophene-*co*-bithiophene) in the direction of the rubbing. A maximum brightness of ≈5,000 cd m^{-2} was reported using these techniques with an EQE of ≈0.13% at the maximum brightness.[31]

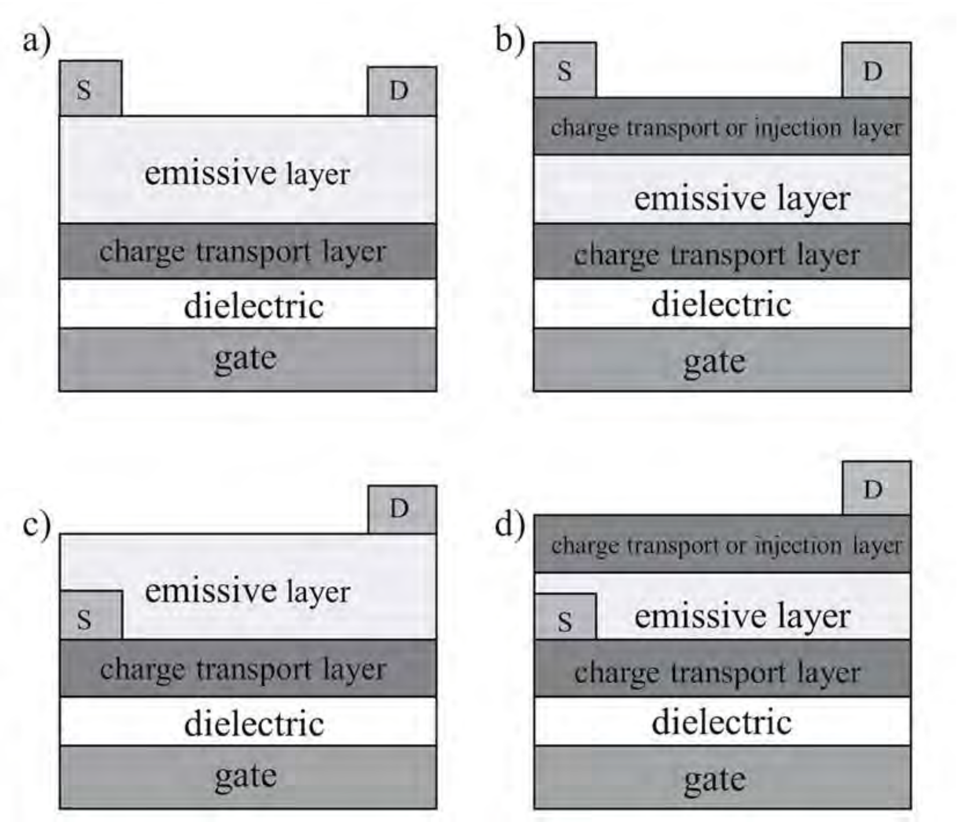

Figure 3. Schematics of **a)** bilayer and **b)** tri-layer LET device top-contact (planar) architectures, and **c)** bi-layer and **d)** tri-layer LETs in a non-planar LET geometry with source (S), drain (D) and gate electrodes. In the bilayer device architecture, a charge transport layer is typically used underneath the emissive layer. The emissive layer is sandwiched between a charge transport layer and either a second charge transport layer or a charge injection layer in the tri-layer architecture.

Tri-layer LETs have also been reported using *n*-type and *p*-type charge transporting materials to sandwich an emissive layer.[32] Electrons and holes were transported in separate layers and recombined in the emissive layer in between. Device performance was dependent on whether the *n*-type or *p*-type layer was closest to the top electrodes. A maximum EQE of 5% was reported but without showing device brightness.[32] A summary of the various multi-layer LETs and their device performance are listed in **Table 2**.

Despite the optical success of the multi-layer all organic LETs, their mobility and ON/OFF ratio remained low. Furthermore, organic-based multilayer structures are notoriously difficult to fabricate, in particular when layers of organic materials have similar solubility, leading to the dissolving or penetration of the bottom layer. To address this issue, a third generation of LETs was developed using organic and inorganic hybrid semiconductor materials. The simplest hybrid approach is to use an inorganic metal oxide as a direct substitute for the organic charge transport layer.[33,34] This already dramatically increases the electron mobility in the resulting hybrid LET and lends the robustness and stability of the inorganic material to the overall device. Complicating the approach slightly with some patterning means that the recombination zone and hence location of the emission from the device can be controlled as well, from a study by Dodabalapur *et al*.[35]

5. Non-Planar Source Drain Contact LETs

It is well known that top contact heterostructures are ideal for charge injection in field effect transistors. This is typically attributed to a better interface with the semiconductor below. A further consideration must be made in LETs due to the ambipolar charge injection of electrons and holes into the device, regardless of its operational polarity. The implication here is that source-drain contact planarity is detrimental to the performance of an LET. Assuming a bilayer LET structure to enhance the electrical performance, the carriers (both electrons and holes) will typically have to be injected and transported through a material. This is best seen in the planar contact devices in **Figure 3b**. Approaching this problem has entailed changing the position or planarity of the contacts, so that the hole and electron injection can occur directly into the hole- and electron-favouring charge transport layers.

An example of the effects of contact placement in organic transistor can be seen in a study by Rost *et al.* that involved a bilayer pentacene/ PTCDI-$C_{13}H_{27}$ architecture.[36] By changing the position of the contacts, the authors could change both the performance and the polarity of the resulting transistor. More relevant to LETs are the studies by Ullah *et al.*[37] and

Muhieddine *et al.*,[33] where the source and drain electrodes were placed in direct contact with the respective charge transport and emissive layers (**Figure 3c** and **3d**). This resulted in a massive drop in contact resistance and an increase in electrical performance that propagated through to the optical performance of the devices. Of significant impact was also the use of asymmetric contacts, *i.e.*, contacts of different materials. In the previously mentioned and other milestone studies,[23] it has been shown that the appropriate selection of electrode materials based on energy level alignment with the charge transport and emissive layers plays an important role in the overall performance of the device.

Table 2. Summary of the various multi-layer LETs and their device performance

Multi-layer LETs							
Ref.	**Device Layout**	**Materials**	μ_{hole} ($cm^2\ V^{-1}\ s^{-1}$)	$\mu_{electron}$ ($cm^2\ V^{-1}\ s^{-1}$)	**ON/OFF Ratio**	**Max Luminance** ($cd\ m^{-2}$)	**EQE (%) at max Luminance**
[38]	2-layers (planar)	DPP-DTT/PDY-132	0.1	0.08	$\approx 10^2$	1,000	2.1
[37]	3-layers (non-planar)	PBTTT/SPB-02T	0.1	0.004	$\approx 10^5$	820	0.05
		PBTTT/SPR-001	0.1	0.0034	$\approx 10^5$	750	0.06
		PBTTT/ PDY-132	0.12	0.003	$\approx 10^5$	2,100	0.06
[39]	3-layers (planar)	PBTTT/MEH-PPV	0.14	–	$\approx 10^7$	112	≈0.001
		PBTTT/PFO	0.16	–	$\approx 10^7$	137	≈0.001
		PBTTT/PDY-132	0.1	–	$\approx 10^7$	647	≈0.001
[40]	3-layers (planar)	PBTTT/PDY-132/NDI	0.05	0.004	–	180	0.0008
[41]	3-layers (planar)	$Ir(piq)_3$	0.42	0.0077	$\approx 10^3$	17.1 µW	3.3
[32]	3-layers (planar)	Alq3-DCM	0.01	0.01	$\approx 10^3$	–	–
[42]	2-layers (planar)	ADS077RE	0.02	3×10^{-7}	$\approx 10^4$	30	0.005
[43]						900	0.25

	2-layers (planar)	DPP-DTT/ PDY-132	0.5	0.2	≈10^3	850	0.09
[44]	2-layers (non-planar)	PBTTT/Dendrimer	0.014	–	≈10^4	650	2.1
[45]	2-layers (non-planar)	DPP-DTT/ PDY-132	7.6	–	≈10^6	29,000	0.4
		DPP-DTT/ PCAN	4.8	–	≈10^5	9,600	0.7
[46]	2-layers (non-planar)	C_{60}/Rubrene	–	0.6	≈10^7	220	0.012
[47]	2-layers (non-planar)	C_8-BTBT/4CzIPN	0.2	–	≈10^6	60	0.034
[48]	5-layers (non-planar)	Pentacene/ TCTA:B3PYMPM	0.11	–	≈10^4	1,890	3.76
		Pentacene/ mMTDAT:OXD-7	0.26	–	≈10^4	1,116	0.93
Hybrid (organic/inorganic) LETs							
Ref. #	**Device Layout**	**Materials**	μ_{hole} (cm^2 V^{-1} s^{-1})	$\mu_{electron}$ (cm^2 V^{-1} s^{-1})	**ON/OFF Ratio**	**Max Power** (W) **/ max Luminance** (cd m^{-2})	**EQE** (%) **at max Luminance**
[49]	2-layers	In_2O_3/ZnO/PDY-132	–	22	≈10^3	700	0.02
[50]	2-layers	IZO/ PDY-132	–	3.5	≈10^7	1,700	0.5
		IGZO/PDY132	–	3	≈10^7	1,800	0.5
[33]	2-layers	ZTO/SPB-02T	–	5	≈10^5	75	0.0001
		ZTO/SPR-001	–	3.7	≈10^5	400	0.0012
		ZTO/PDY-132	–	4.2	≈10^4	400	0.0018
[51]	2-layers	ZTO/PDY-132	–	1.62	≈10^5	1,330	0.087
		ZTO/F8BT	–	0.45	≈10^6	226	0.038
		ZTO/ PFO	–	0.38	≈10^4	150	0.07
[52]	2-layers	AZO/BP3T	–	0.01–0.001	–	–	–
[53]	1-layer (blend)	ZTO/Tetracene	–	0.81	–	–	–

[54]	2-layers	ZnO/Tetracene	–	14	–	–	0.000067
[34]	2-layers	CdS/PDY-132	–	19.5	$\approx 10^{7}$	2,100	0.01
[55]	2-layers	ZTO/Pt(II) complexes	–	1.3	$\approx 10^{7}$	744	0.082

6. LET Materials

Selecting organic semiconducting materials that meet the requirements for use in an LET architecture is not trivial as suitable material combinations must facilitate balanced charge injection, transport and recombination in order to achieve high device performance. Interestingly, simply reusing already established OLED materials does not necessarily give rise to successful LET devices. To understand what determines the suitability of LET materials, one has to consider that generally high charge mobility and light emitting properties are contradicting parameters – but both need to be implemented into a LET. The reason behind this inherent trade-off is that high mobility materials provide large intermolecular overlaps over highly ordered layers. In turn, device EQE is significantly reduced due to such aggregation induced self-quenching of photoluminescence. This phenomenon becomes obvious for Hepp's reported first light-emitting transistor based on the unipolar tetracene.[4] Tetracene offered reasonable hole mobilities (10^{-2} cm^{2} V^{-1} s^{-1}) from its highly ordered films but only delivered poor light output. Although quite dim, this first LET paved the way towards much more advanced and effective layouts in which more bespoke materials have been pushing LET technologies to ever higher performance. Over the years, it has become apparent that an ambipolar LET design that utilises materials exhibiting hole and electron transporting properties is advantageous, in that it allows balanced charge injection and transport into the emissive layer.

In this section we will first introduce common electrode materials, followed by a selection of active organic materials used in contemporary charge injecting and emissive layers. As mentioned *vide supra*, finding a single material that simultaneously provides appropriate charge transport and emissive properties is nigh on impossible. Thus, we will distinguish between charge injecting materials and organic emitters and aim to aid the reader selecting matching material combinations for their envisioned devices.

6.1 Electrode Materials

There are a large variety of electrode materials that can be chosen from. These materials are usually metals that are vacuum deposited on the device. It is important to note that despite of the ease of electron injection, most low work function metals are prone to oxidation and hence require measures to prevent air exposure. Metal oxides (*e.g.*, ZnO) on the other hand has been used as an air-stable electron-injecting electrode in ambipolar LETs.[56] It is also worth mentioning that by proper design of the thickness and refractive index, semi-transparent electrodes can be accomplished. This potentially allows the use of LETs in next-generation semi-transparent display with pixel aperture ratios exceeding those of active matrix organic light-emitting diodes (AMOLEDs).[57]

6.2 Charge Transporting Materials

Similar to electrode materials, charge transporting materials are generally classified in either electron or hole transporting for unipolar devices, as well as ambipolar materials for devices as noted previously. The latter is quite desirable as fewer device layers are required, compared to unipolar layouts.[58] However, as charge mobility is a major requirement for LETs, thus far ambipolar charge transport materials are struggling to deliver the needed performance in that regard.

Historically, acenes such as tetracene and pentacene (**Figure 4**) have been a promising charge transporting materials. Their extended conjugated systems along with the tendency to form highly crystalline layers provide excellent intermolecular interaction and consequently high charge mobility. Prominent materials such as tetracene and rubrene have been widely used in the field.[4,59] Hole mobilities of close to 2 cm^2 V^{-1} s^{-1} reported underlined the advantage of highly ordered film for charge injection. However, this macromolecular arrangement is only accessible by vacuum deposition. Moreover, the π-fused acenes are inherently susceptible to oxygen and moisture, which limits their practical application in terms of long-term stability. This is especially true for tetracene and hence motivated Dadvand *et al.* to combine the less pronounced π-system of anthracene with a phenylvenylne moiety to increase stability whilst maintaining the high hole mobility of larger acenes (2.6 cm^2 V^{-1} s^{-1} in single crystals and 1.5 cm^2 V^{-1} s^{-1} in thin films).[60] This material also gave a high solid state PLQY of 70% and was used as a blue emitter in a unipolar LET.[60] The authors attributed the high mobility and high PLQY to the high energy of the excited state T_1 relative to S_1 ($2T_1 > S_1$), which turns off the emission quenching through singlet fission.

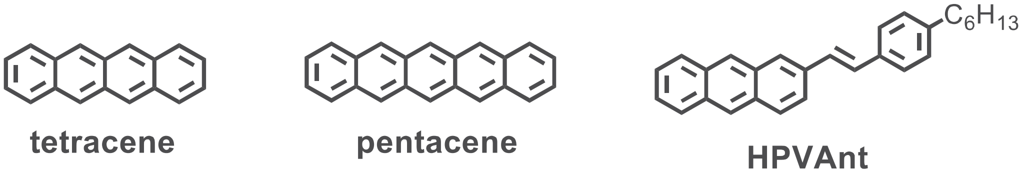

Figure 4. Chemical structures of acene derivative examples.

BTBT (benzothieno[3,2-b][1]benzothiophene) and its derivatives (see **Figure 5**) have been demonstrated as a most promising class of materials for LETs as well as transistors, organic solar cells and photodetectors.[26,47,61,62] Due to their unique macromolecular arrangements, their fused benzothiophene cores enable strong orbital coupling between adjacent molecules. When additionally equipped with long alkyl chains, BTBTs create layers, in which charge transport predominantly occurs perpendicular to the molecules' π-system via a 'hopping' mechanism.[61] For their remarkably high hole mobility properties (generally >1 cm^2 V^{-1} s^{-1}), their transparency in the visible spectral range and their capability of operating in LETs at low voltages (<10 V), they are rightfully subject of intense contemporary research.[57,62]

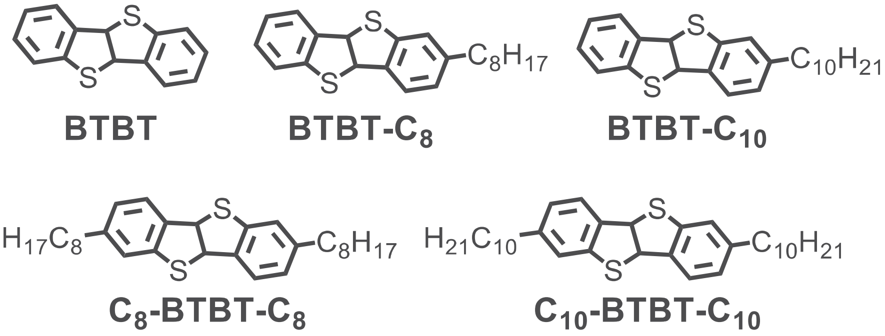

Figure 5. Chemical structures of hole transporting BTBT examples with high hole mobilities.

As mentioned before, one can find fewer *n*-type than *p*-type organic semiconductor materials. This renders novel and innovative electron-transporting materials quite desirable as balanced charge injection is a major requirement for many LET devices. To achieve *n*-type charge transport, inorganic semiconductors such as CdS [34] and metal oxides (*e.g*., ZnO, ZTO) have been developed to offer a much higher charge mobility of up to 19 cm^2 V^{-1} s^{-1} and a high channel current ON/OFF ratio (>10^6).[49-51] However, the major drawback of inorganic charge transporting materials is their very high processing temperatures (typically >500 °C).

6.3 Fluorescent Emitters

The common active light-emitting materials used in organic LETs have been mainly based on fluorescent emitters. A wide range of fluorescent emitters used in efficient OLEDs have been studied in organic LETs. This has included polyfluorene and phenylene-vinylenes based conjugated polymers. Polyfluorenes based polymers are quite unique due to their ambipolar charge transport capabilities and high photoluminescence quantum yield.

Furthermore, they can be chemically modified to emit light throughout the visible range: F8, F8BT and F8TBT (**Figure 6**) were used as ambipolar materials in single and mixed layer devices to give blue to red emission.[63-65] Kajii *et al.* demonstrated a blue emitting device where differently orientated layers of F8 improved hole and electron injection.[66] They also showed green phosphorescent emission from $Ir(piq)_3$-doped in F8. Mobilities of polyfluorene-based devices are usually in the range of $10^{-3} – 10^{-4}$ $cm^2 V^{-1} s^{-1}$.

Phenylene-vinylenes based conjugated polymers were the materials used in the first solution based LETs.[12] They are commonly used in ambipolar devices – mostly as charge transporting and emitting material concurrently. Here prominent derivates are OC_1C_{10}-PPV and Super-Yellow.[67,68] However, the LET characteristics from singlet layer fluorescent emitter have been far from practical. As aforementioned, the efficiency and brightness in single layer LETs closely relate to the matching combinations of charge injecting electrodes and materials energy levels. This evidently also holds true for the way an emitter is positioned in the device. Emissive layers sandwiched between the hole and electron injecting layers are amongst the most common layouts. The incentive here is – given a balanced charge injection - to achieve efficient recombination in the emissive layer.

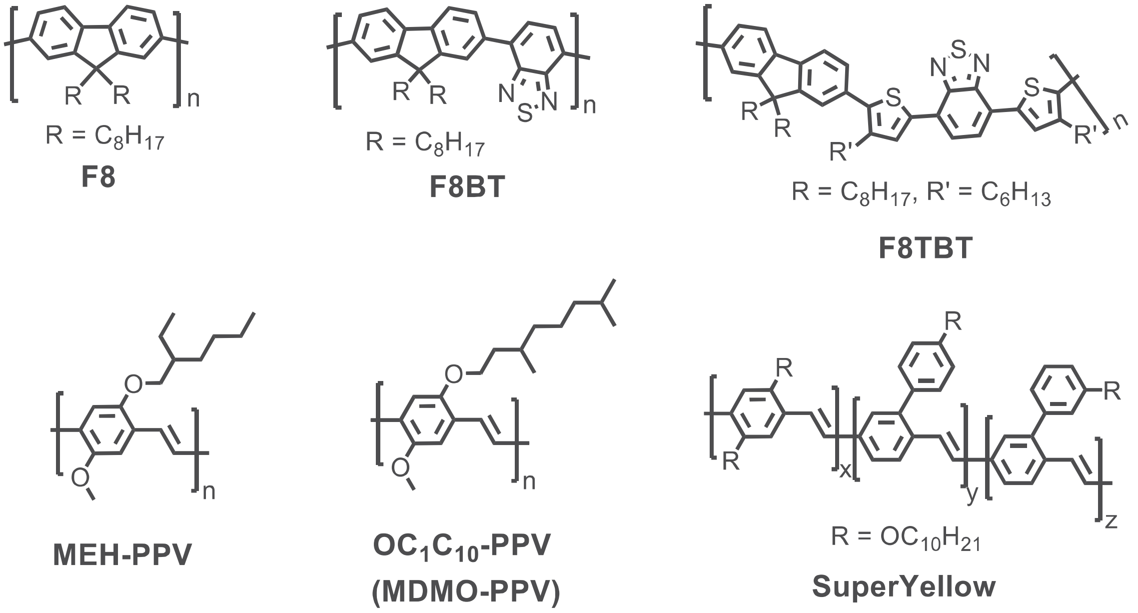

Figure 6. Chemical structures of fluorescent light-emitting polymers examples.

6.4 Phosphorescent Emitters

The majority of materials used in LETs have based on fluorescent semiconductors. One major drawback they commonly share is that they only exploit the recombination from singlet excitons. This leaves up to 75% of the input energy (*i.e.*, triplet excitons) wasted as non-radiative decay, leading to poor device performance. In contrast, the use of phosphorescent materials allows for harnessing both singlet and triplet excitons generated in LETs due to efficient inter-system crossing, rendering them advantageous to their fluorescent counterparts. Many phosphorescent OLED materials have been based on precious metal complexes such as iridium and platinum. Representative for the metal-complex family especially iridium-based emitters have been explored in LETs. For example, in 2009 Namdas *et al.* presented solution-processable dendrimers (*e.g.*, IrG1 and IrG2) in CBP, 4,4′-bis(*N*-carbazolyl)-1,1′-biphenyl, host to give green organic LETs as well as red emitting LETs while using ADS077RE (**Figure 7**).[42] Phosphorescent red LETs have been further demonstrated by exploiting $Ir(piq)_3$ as the emitter.[42,69,70] Similar to $Ir(piq)_3$, another very well-known player from OLED research has found its way into organic LETs of late – the green emitter, $Ir(ppy)_3$.[48,71,72] So far, the device EQEs of these phosphorescent LETs are far lower than their OLEDs, which can be at least in part attributed to their low charge carrier mobility and poor recombination of these phosphorescent emitters in the host despite of the use of high hole transporting polymer (*e.g.*, PBTTT) underneath (*i.e.*, a bi-layer device structure). To overcome the low charge mobility of these phosphorescent emitters in host, a host free blue dendrimer was studied to achieve a good mobility ($1.4 \pm 0.5 \times 10^{-2}$ cm^2 V^{-1} s^{-1}) and EQE (2.2±0.5%) with high brightness from standard electrode-edge emission. In contrast to octahedral Ir(III) complexes, square-planar Pt(II) complexes have been less investigated in organic LETs but might have greater potential for their 2D structure in nature that allows for strong intermolecular interaction in achieving potential higher charge carrier mobility and potentially high light-output coupling. More importantly, many Pt(II) complexes have lately been demonstrated to exhibit unique aggregate-induced phosphorescence enhancement (either through MMLCT transition or restriction of rotation in neat films)[73,75] instead of aggregate-caused quenching in most Ir(III) complexes. While neat films of Pt(II) complexes with aggregate-induced phosphorescence enhancement are yet to be demonstrated in organic LETs, full electrode emission has been shown for LETs based on a red Pt(hopmq)(acac) blend film, in addition to a high electron mobility of 1.3±0.1 $cm^2 V^{-1} s^{-1}$ and ON/OFF ratio of $4\pm1 \times 10^6$.[55] Although, phosphorescent emitters have a great potential in push exciton-light efficiency further, they also have a key drawback, that is often expensive for these precious metals.

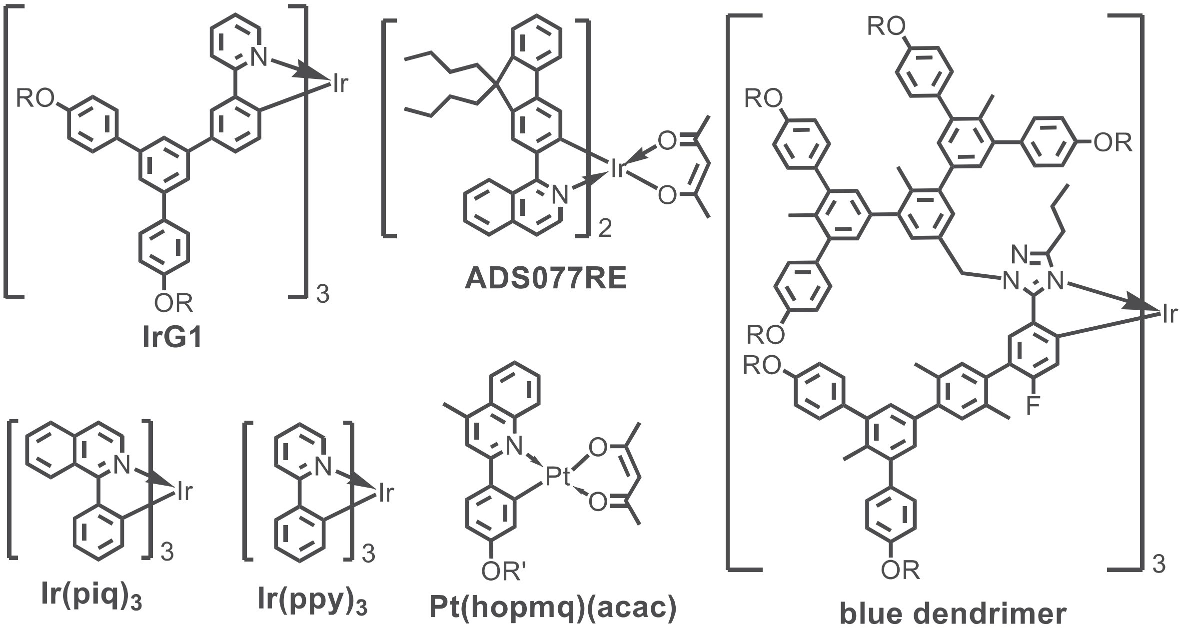

Figure 7. Chemical structures of phosphorescent light-emitting material examples used for LETs; R = 2-ethylhexyl and R' = *n*-hexyl.

6.5 TADF Emitters

Recently, an alternative solution to triplet harvesting and expensive metals problems has appeared in the form of thermally activated delayed fluorescence (TADF) materials.[76] Molecules exhibiting this phenomenon are carefully engineered to have a small energy gap between their singlet and triplet. Hence, the thermal energy (coming from room temperature) is sufficient to enable efficient reverse intersystem crossing of triplets into the singlet manifold, followed by their fast, radiative decay. Even though the whole process is not fully understood yet – with more factors than simply energy level difference coming into the picture[76-80] – several highly efficient OLEDs have been reported, covering different parts of the visible spectrum.[77] To date, there are only few reports of usage of this class of materials in organic LETs. The first report describes devices based not on the TADF capable single molecule, but a thermally activated TCTA:B3PYMPM donor-acceptor system.[48] There, the authors showed a modest EQE (close to 1%) and brightness exceeding 1,000 cd m^{-2}. However, the first report of LETs using true intramolecular TADF material was the one utilizing 4CzIPN.[47] Both p- and n-type transistors were presented with an EQE reaching 0.1% and brightness (see **Figure 8**) in the range of 100–1,000 cd m^{-2} (depending on the polarity and type of electrodes used). The low EQE has been mainly attributed to the recombination zone being under the metal electrode. Given the promise of almost unitary internal quantum efficiency of TADF materials, more work should be followed in the near future.

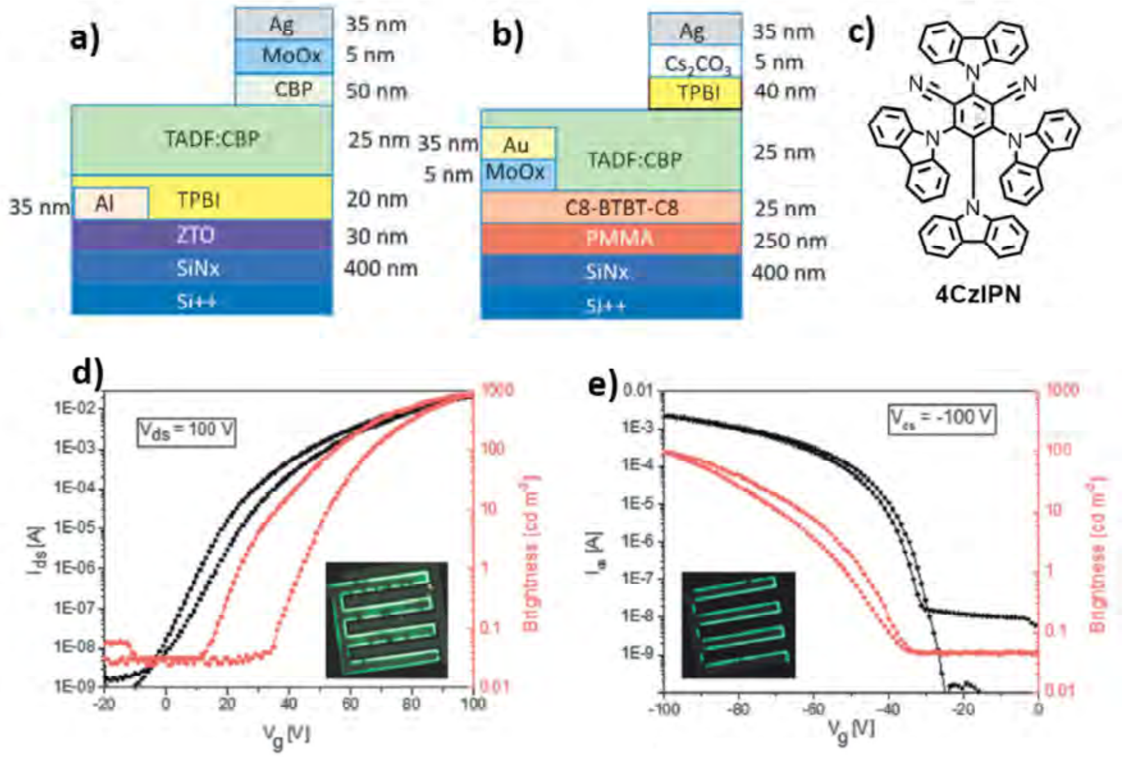

Figure 8. Device structures of n–type (**a**) and p-type LETs (**b**) based on 4CzIPN TADF (with chemical structure in **c**). Current-voltage and Luminesce characteristics of LETs (**d** & **e**). Reproduced with permission.[47] Copyright 2018, Wiley-VCH.

6.6. Perovskite Based Emitters

Recently, metal-halide perovskite materials have shown a great promise for optoelectronic applications including light-emitting transistors.[81-87] Although metal-halides have been shown extremely high charge carrier mobilities (up to 3,100 cm^2 V^{-1} s^{-1}) in theoretical calculations, the first report to metal-halide perovskite based LET came out in 2015 using methylammonium lead iodide ($MAPbI_3$). In this report, Chin *et al.*[87] utilised a pre-patterned bottom-gate, bottom contact transistor architecture and active layer of $MAPbI_3$ was deposited as a final fabrication step to minimize the effects of degradation of the active material caused by fabrication steps from environments. Temperature activated transistor behaviour was observed below 198 K with an increasing ambipolar character at lower temperatures. The hole and electron mobilities at 78 K were 6.7 × 10^{-2} cm^{-2} V^{-1} s^{-1} and 7.2 × 10^{-2} cm^2 V^{-1} s^{-1}, respectively, extracted from the saturation regime. Electrode emission was observed, when the LET was operated at a temperature range of 78–178 K, with 178 K being the maximum temperature at which the light-emission could be observed. The nature of the activated hole and especially electron transport in reported LET structure limits the use of these devices at room temperature. Subsequently, a top contact perovskite LET, allowing room temperature operation and light emission in AC driving mode at high-frequency modulation of the applied voltages, was demonstrated by Maddalena *et al.*[88,89] The high frequency operation suppressed the ionic drift/screening in the perovskite.

Recently, solution processed perovskite LETs[90] using two classes of materials in a heterostructure configuration (see **Figure 9**) were demonstrated, i) self-organized NFPI7 MQW perovskite as the light-emitting layer, and ii) high electron mobility In_2O_3/ZnO as the charge transporting layer. The combination yielded relatively efficient (0.2% EQE at 18 cd m^{-2}) LETs operated at room temperature, with a high electron mobilities approaching to 21 cm^2 V^{-1} s^{-1}. An important feature of this study is that a large and uniform near infra-red emission (with peaks at 783 nm) was observed under the entire drain electrode region (hole injecting contact). The overall electrical and optical performance of the heterostructure LETs are orders of magnitude better than previously reported, which opens up further research opportunities towards development of a variety of solution processed hybrid optoelectronic devices.

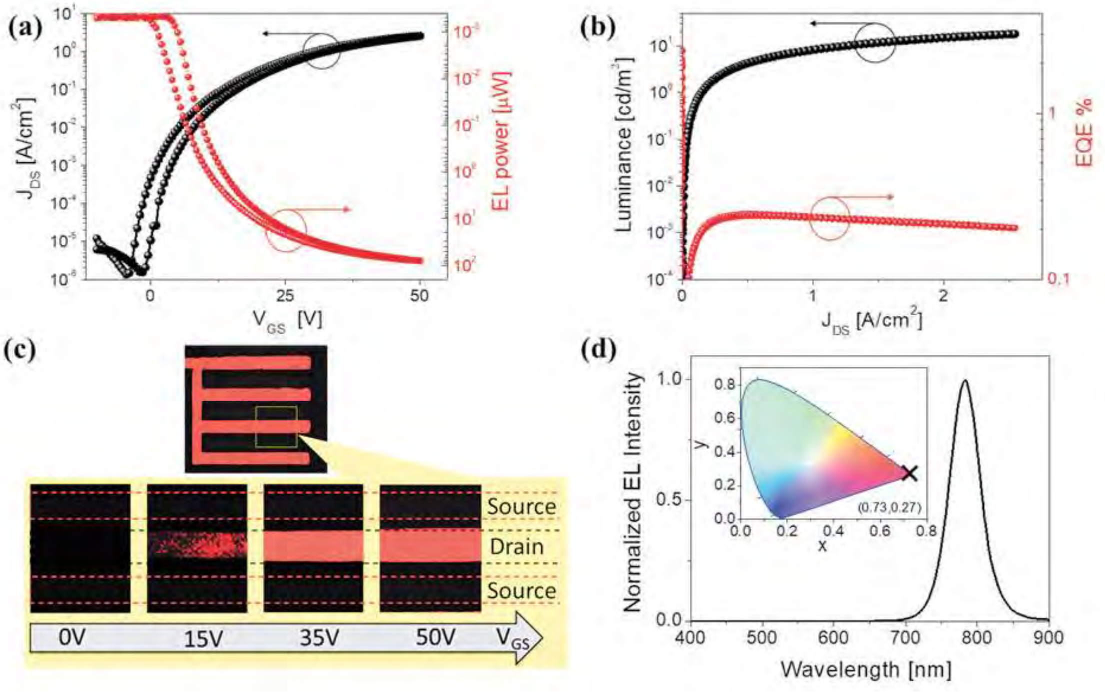

Figure 9. **a**) Typical I-V-L characteristics for a NFPI7-based LETs; **b**) luminance and EQE (%) plotted versus current density; **c**) optical microscopy images of the LET during operation; **d**) EL spectrum and the Commission Internationale d'Eclairage (CIE) color coordinates (0.73, 0.27) of the LET. Reproduced with permission.[90] Copyright 2019, Wiley-VCH.

7.0 Low Operating Voltage LETs

LETs provide gate controlled light emission and channel conductivity, in a similar fashion to OLEDs with thin films transistors backplane. Therefore, in order to obtain high drain current at lower operating voltages, several key parameters play the roles and need to be considered; i) charge carrier mobility in active layer, ii) dielectric capacitance of the dielectric film, iii) interfacial and bulk trap states and iv) charge injection. The intrinsic lower mobility combined with lower dielectric capacitance in most of organic LETs reported leads to high operating voltages (≈100 V) for best performance, hindering the demonstration of power-efficient devices. A simplified relation of transistor drain current in saturation regime is given below:

$$I_{DS} = \frac{1}{2} \times \frac{W}{L} \times \frac{\varepsilon\,\varepsilon_o}{d} \times \mu_{FE} \times (V_{GS} - V_{TH})^2$$

where I_{DS} is drain current, W/L is channel width to length ratio, ε is permittivity of free space, ε_o is dielectric constant of the dielectric material, d is dielectric thickness, μ_{FE} is charge carrier mobility, V_{GS} is gate voltage and V_{TH} is threshold voltage. In order to lower the operating gate voltages, higher charge carrier mobility materials are requisite. However, most of highly luminescent organic light-emitting materials suffer from poor charge mobility. Therefore, to overcome the intrinsic lower mobility, heterostructure LETs have been reported utilizing an organic emissive material in combination of a wide range of high mobility materials including polymers,[37-38] small molecules,[26,32] metal oxides,[49-51] and carbon nanotubes (CNTs).[91]

Another solution to lower the operating voltages is to increase the capacitance of the dielectric film. This can be achieved by lowering the thickness of the dielectric films and/or through the use of high-k dielectric materials. Lowering the film thickness is challenging in order to produce, pin-hole free, smooth dielectric films. However, recent reports suggest that atomic layer deposition (ALD) and solution processing techniques have been effective to fabricate low voltage LETs. A combination of high dielectric materials and lower film thickness can be most effective. Recently, Mujeeb *et al.* have shown that a combination of high-k and low-k dielectrics based 2-layer dielectric strategy was effective to lower the operating voltage below 10 V by reducing the charge trap states at dielectric/charge transport layer.[49] McCarthy *et al*. employed a thin layer of ALD grown Al_2O_3 capped with a BCB dielectric layer in combination with high mobility CNT charge transporting layer to significantly lower the operating voltage of LET.[91] This resulted in high electroluminescence efficiency in LET with reduced power dissipation.

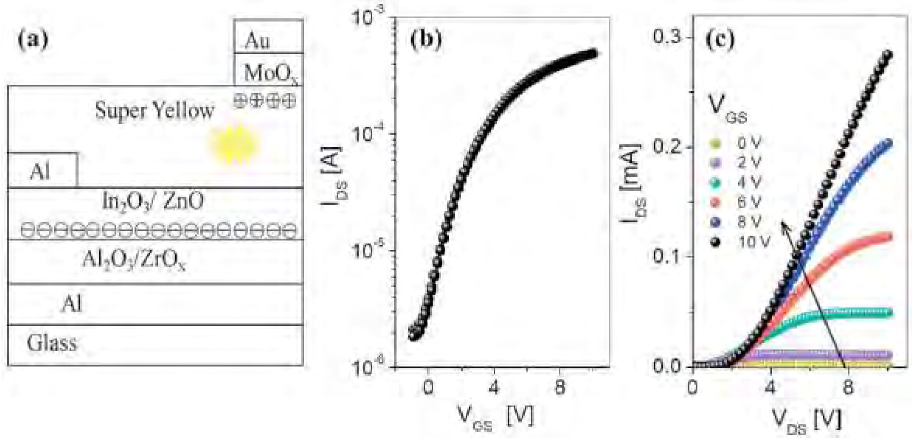

Figure 10. Schematics of low voltage operating oxide based solution processed LET (**a**), and the electrical characteristics (**b** & **c**). Reproduced with permission.[49] Copyright 2018, American Chemical Society.

8. Outlook

Currently, there is a huge demand for transparent display technology as it opens up new markets like augmented reality, automobile navigation systems, car windscreen displays, and head mounted goggles for biomedical or surgical applications. To realise this next step in display technology, highly transparent display pixels with ultra-high brightness are required. The existing state-of-the-art AMOLED pixels have been shown to be ineffectual. Therefore, there is an urgent need to develop a new class of transparent display pixels.

LETs offer a new route to remarkably simplified display pixels as they directly provide the light-emitting function of an OLED with the switching capability of field-effect transistors (FETs) in a single device architecture. Such an integrated pixel has the potential for high transparency, low-cost and simple fabrication as it requires less circuitry than what is usually involved in conventional AMOLED pixels. In our recent proof-of-principle study, we have demonstrated that an LET pixel can deliver unprecedented aperture ratio of ≈80%[51] (defined as the ratio of light-emitting area over total pixel area), easily surpassing the minimum requirement for AMOLED displays. However, significant advances still need to be made before LETs can truly be incorporated into displays or transparent display pixels as a viable alternative to the existing AMOLEDs.

As discussed earlier, the current main issue is poor light outcoupling from underneath the source and drain contacts. Tuning the transparency of the metal contacts used would allow significant improvement of light outcoupling of the devices at the same current and applied voltage. The operating voltage of most LETs are still inhibitively high. Even in the multi-layer configuration, hundreds of volts are needed to achieve the brightness that would be required for display applications. One possible route towards overcoming this would be the use of high k gate dielectrics. High k dielectrics such as aluminium oxide have been shown as a viable option for low operating voltage OFETs.[57]

For practical application, device stability is important. Studies into continual bias stress testing with the application of applied voltages to date have not been reported for LETs. This means that it is unknown how suitable these devices could be for a working display. Finally, from a material side, the development of a new class of LET compatible materials is urgently needed. The ideal LET materials would display moderate charge transport, a high PLQY and less EQE roll-off.

LETs have long been an attractive architecture in the development of organic injection lasers thanks to their potential to achieve high current densities[27,92] and a well-defined recombination zone, far from loss-inducing metal contacts. However, since the infamous paper by Schon on false injection lasing claim in organic materials, the scientific community is still cautious when it comes to newly appearing lasing claims. In order to achieve injection lasing, the light-emitting organic materials should ideally have high mobility (in order to achieve high current densities easily) and quantum efficiency (in order to have a chance of achieving net gain) at the same time, properties that are usually mutually exclusive in organic materials.

Ambipolarity is also desirable in order to mitigate losses coming from hole and electron current mismatch.

Interesting strategies addressing this problem have been developed recently by introducing LETs with a split-gate[17] and overlapping split gate structures,[93] where hole and electron currents can be controlled independently. These structures provide high EQEs and finer control over the recombination zone. In order to achieve lasing, the active material in an LET should also avoid other loss pathways introduced by injection pumping, namely polaron losses (coming both from polaron absorption[94,95] and polaron quenching[96] and excited state or triplet absorption (which should lie outside of the singlet emission wavelength). Finally, the long triplet lifetime[97] and spin statistics of exciton generation lead to the very rapid pile-up of triplet population. It is therefore important to pump the device with nanosecond scale electrical pulses, with large intervals between pulses.[98] Despite all those obstacles, a recent report has been published on injection organic lasing using BSBCz in diode structure.[99] Nevertheless, LET architecture is thus far the promising pathway to unlock this phenomenon.

Acknowledgements

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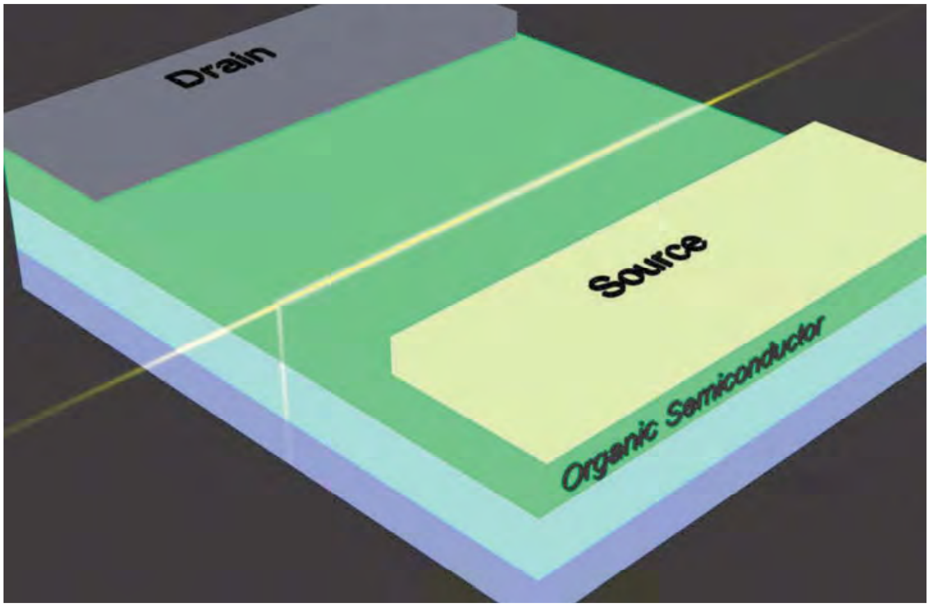
TOC image



Light emitting transistors (LETs) is a multifunctional device that combines the switching abilities of a transistor with the emissive properties of an LED. This progress article focuses on the state-of-the-art development of LETs and its prospective for the potential applications in transparent displays and organic injection lasers.

A passport-type photo and a short summary of the corresponding authors

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