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# Charge transport and recombination in heterostructure organic light emitting transistors

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ABSTRACT

Light-emitting field effect transistors (LEFETs) are a class of organic optoelectronic device capable of simultaneously delivering the electrical switching characteristics of a transistor and the light emission of a diode. We report on the temperature dependence of the charge transport and emissive properties in a model organic heterostructure LEFET system from 300 K to 135 K. We study parameters such as carrier mobility, brightness, and external quantum efficiency (EQE), and observe clear thermally activated behaviour for transport and injection. Overall, the EQE increases with decreasing temperature and conversely the brightness decreases. These contrary effects can be explained by a higher recombination efficiency occurring at lower temperatures, and this insight delivers new knowledge concerning the optimisation of both the transport and emissive properties in LEFETs.



## 1. Introduction

Organic light-emitting field effect transistors (LEFETs) are dual function devices in that they have the electroluminescence capabilities of organic light-emitting diodes (OLEDs) plus the switching capabilities of a field effect transistor (FET) in a single architecture [1–5]. This dual functionality can potentially lead to new applications such as simplified pixels for flat panel displays and potentially an electrical driven organic semiconductor laser. Although the emission brightness of LEFETs has improved over the last decade [6–17], their electrical switching ON/OFF ratio and EQE at high brightness are still very much sub-optimal. This is in part due to a lack of knowledge with respect to materials design and the absence of a comprehensive understanding of the charge transport and radiative recombination processes that occur in an LEFET.

A common method to probe the charge transport in organic semiconductor films is to measure the optoelectronic characteristics of the material as a function of temperature in an OFET or diode configuration [18–24]. Specifically, in the diode configuration both transient (e.g., Time-of-Flight [25], photo-Charge Extraction in Linearly Increasing Voltage [photo-CELIV] [26]) and steady-state measurements (e.g., Space Charge Limited Current [SCLC]) have been used to measure mobility and recombination. Recently, Armin et al. reported an adapted injection-CELIV technique called MIS-CELIV which is capable of measuring the mobility of both carrier types in diode architectures relevant to operational devices such as solar cells and photodiodes [27]. However, in a functional OLED, we need to simultaneously probe not only the transport properties, but also the recombination dynamics (radiative and non-radiative). Thus, the traditional transport measurement methodologies only uncover at best half the pertinent physics. Furthermore, these existing techniques require that the injecting contact must be Ohmic. Such a requirement is hard to achieve in an organic diode configuration. In an OFET architecture, the contact resistance at the organic-metal interface can be completely eliminated by employing four-probes or the transmission line technique [28–31]. An OFET structure can potentially map multiple elements of transport such as charge injection at the organic-metal interface, contact resistance and mobility. However, OFETs are generally not designed to emit light and are thus, like the simple diode, not suitable for studying radiative and non-radiative recombination processes.

In this work, we simultaneously probe the mobility and injection of carrier types, contact resistance and radiative recombination all as a function of temperature in a model bilayer LEFET comprised of a light-emitting and a charge transporting polymer. The bilayer LEFETs show decreases in the source–drain current, mobility (both electrons and holes), and brightness with decreasing the temperature. However, the external quantum efficiency

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(EQE) increases by an order of magnitude at low temperatures. The changes in mobility and current are thermally activated consistent with the hopping transport characteristics normally associated with disordered semiconductors. We thus demonstrate that the increase in EQE at low temperature is predominately due to an increase in the radiative recombination efficiency.

## 2. Experimental procedure

### 2.1. Device preparation

Fig. 1a shows the device architecture for top-contact, bottom-gate LEFETs. The devices were fabricated on 400 nm thick SiNx gate dielectric layers, grown by low pressure physical vapour deposition (LPCVD), on top of a heavily n-doped silicon wafer purchased from Silicon Quest, International, Inc. After dicing the wafer into $15 \times 15$ mm substrates, they were cleaned with ultra-sonication in acetone for 20 min, followed by ultra-sonication in 2-propanol for 20 min. The substrates were then dried with nitrogen. All remaining fabrication steps and device testing were performed inside a nitrogen filled MBraun glove box ($O_2$ and $H_2O$ levels <0.1 ppm). The gate dielectric layer was further passivated with poly(methylmethacrylate) (PMMA) (120,000 gmol). A 35 mg/ml solution of PMMA in *n*-propylacetate (P99.5%) was spin-coated onto the substrates at 2500 rpm for 30 s and then 3000 rpm for 10 s. The substrates were then baked on a hot plate at 150 °C for 30 min. The PMMA film thickness was 120 nm. The materials used for the semiconducting channel were: an ambipolar diketopyrrolopyrrole–dithienothiophene (DPP–DTT) co-polymer [32]; and Super Yellow (SY), a phenyl-substituted poly(*p*-phenylenevinylene) co-polymer, as the emissive layer. SY (PDY-132) was purchased from Merck and was used without further purification. The reported chemical structures for both these polymers are illustrated in Fig. 1(b and c). The DPP–DTT layer was deposited on top of the PMMA layer from a 4 mg $mL^{-1}$ solution in chloroform with 7% of 1,2 dichlorobenzene (⩾99%, anhydrous). In order to fully dissolve the DPP–DTT polymer, the solution was heated at 80 °C followed by a slow cooling protocol [33] and spin-coated at 1000 rpm for 60 s. This step was followed by baking the sample on a hot plate at 150 °C for 30 min. A 7 mg $mL^{-1}$ solution of SY in toluene (>99.9% anhydrous) was spin-coated on top of the DPP–DTT layer at 2500 rpm for 30 s then at 3000 rpm for 10 s. The substrates were then annealed on a hot plate at 150 °C for 30 min. The LEFETs were completed by deposition of asymmetric electrodes under vacuum ($2 \times 10^{-6}$ mbar) through shadow masks prepared by deep reactive ion etching with channel widths of 16 mm and four different channel lengths 50, 80, 100 and 120 μm. Asymmetric source and drain contacts were deposited in two separated evaporations, first Au (hole injection) and then Ba (electron injection). The two metals were chosen due to their work functions being good matches for the relevant semiconductor energy levels to ensure optimised charge injection [17]. Devices were encapsulated with Cytop CTL-809 M (solvent: CTSolv.180) from Asahi Glass Japan.

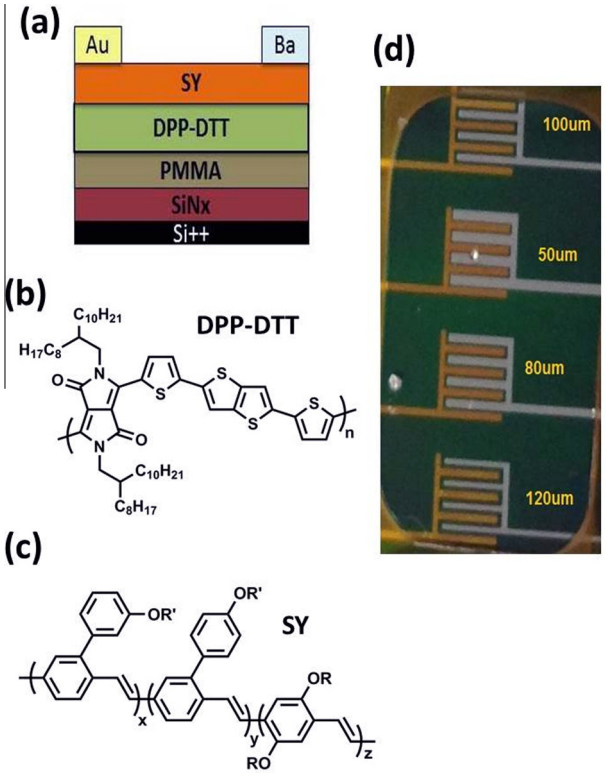


**Fig. 1.** Schematic LEFET device architecture, chemical structures, and optical images of the LEFETs used in this study: (a) LEFET device architecture using asymmetric source and drain contacts; (b) chemical structure of DPP-DTT; (c) chemical structure of SY; (d) optical images of LEFETs with different channel widths on one substrate.

### 2.2. Temperature dependent measurements

A Janis closed-cycle Helium Opti-Cryostat was used for the temperature dependent measurements from 300 K to 135 K. After mounting the LEFETs onto the cryostat finger, the sample chamber was evacuated ($5 \times 10^{-5}$ mbar) and refilled with He. The devices were cooled down from ambient and at each temperature, transfer and output characteristics, and emission intensity were recorded simultaneously for both electron and hole accumulation modes. Electrical characteristics of the devices were acquired using an Agilent B1500A Semiconductor Device Analyser at each temperature. The emission was recorded as photocurrent with a calibrated photomultiplier tube (PMT) positioned at one of the optical windows of the cryostat. The brightness was calculated from the PMT photocurrent by comparison with a device with known brightness [15]. The EQE was calculated from the ratio of emitted photons to the number of injected charge carriers, which were extracted from the brightness and source–drain current, respectively, assuming Lambertian emission as described by Greenham et al. [34].

The charge carrier mobility ($\mu$) was calculated in the saturation regime ($V_{DS} \geqslant V_G - V_t$) of the *I–V* curves using the MOSFET equation: [35]

$$\mu_{sat} = \left(\frac{2L}{wC_i}\right)\left[\frac{\partial\sqrt{I_{DS}}}{\partial V_g}\right]^2, \quad (1)$$

where $C_i$ is the insulator capacitance per unit area, $w$ and $L$ are channel width and length, respectively, $I_{DS}$ is the source–drain current, and $V_g$ is the gate voltage. The gate capacitance is comprised of the SiNx and PMMA layers, which were estimated as the sum of the capacitors in series.

The LEFET shadow masks were designed to have four devices on each substrate with the same channel width but different channel lengths as shown in Fig. 1d. In order to extract the intrinsic mobility, the total resistance was deduced from the output characteristics at different temperatures and for at least three devices with different channel lengths on one substrate. The contact resistance was extracted by extrapolating the intercept for a channel length equal to zero. Intrinsic mobilities were calculated as per standard procedures [36–40].