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Published in:
Applied Physics Letters

DOI:
10.1063/1.1785290

Publication date:
2004

Citation for published version (APA):
Rost, C., Karg, S., Riess, W., Loi, M. A., Murgia, M., & Muccini, M. (2004). Ambipolar light-emitting organic field-effect transistor. Applied Physics Letters, 85(9), 1613 - 1615. https://doi.org/10.1063/1.1785290

APPLIED PHYSICS LETTERS VOLUME 85, NUMBER 9 30 AUGUST 2004

Ambipolar light-emitting organic field-effect transistor

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(Received 31 March 2004; accepted 22 June 2004)

We demonstrate a light-emitting organic field-effect transistor (OFET) with pronounced ambipolar current characteristics. The ambipolar transport layer is a coevaporated thin film of α-quinquethiophene (α-5T) as hole-transport material and N,N′-ditridecylperylene-3,4,9,10-tetracarboxylic diimide (P13) as electron-transport material. The light intensity is controlled by both the drain–source voltage V_{DS} and the gate voltage V_G. Moreover, the latter can be used to adjust the charge-carrier balance. The device structure serves as a model system for ambipolar light-emitting OFETs and demonstrates the general concept of adjusting electron and hole mobilities by coevaporation of two different organic semiconductors. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1785290]

Organic materials have been incorporated as active layers in electronic thin-film devices, such as organic light-emitting diodes (OLEDs),[1,2] organic solar cells,[3,4] electrochemical cells[5] and organic field-effect transistors (OFETs).[6–9] The progress in the field of OLEDs for display applications was recently highlighted by the demonstration of a 20-inch full-color active-matrix OLED display driven by amorphous Si thin-film transistors.[10] OFETs are being developed as switching devices for active-matrix OLED displays[11] and for low-cost electronics, such as low-end smart cards and electronic identification tags. Combining optical and electrical functionality in a single device, i.e., a light-emitting field-effect transistor (LEFET), would not only increase the number of potential applications in integrated circuitry for signal processing that involves both optical and electrical signals, but also present an ideal structure for lifetime studies of organic light-emitting materials under different driving conditions and charge-carrier balances. Recently, a unipolar light-emitting OFET based on tetracene was reported.[12–14] However, in unipolar devices, light emission is restricted to a region very close to the contact that injects the charge carriers of the lower mobility. In contrast, an ambipolar light-emitting transistor would allow the electron–hole balance as well as the location of the recombination zone between source and drain electrodes to be tuned by the gate voltage, hence improving the quantum efficiency. In principle, an ambipolar transistor without light emission can be formed by using either a material capable of transporting both electrons and holes or a heterostructure consisting of a hole- and an electron-transport material. The latter has been demonstrated for combinations of α-hexithienylene (α-6T) and C_{60} (Ref. 15) as well as of pentacene and N,N′-ditridecylperylene-3,4,9,10-tetracarboxylic diimide (P13).[16] Ambipolar transport in a wide-band-gap organic material, necessary for light emission in the visible region, is difficult to achieve because of impurity-induced traps.[17] A viable way to circumvent this problem is to mix electron- and hole-transporting moieties into one phase, as was demonstrated with solution-processed OFETs in Refs. 18 and 19.

Here, we report on field-effect transistors based on a coevaporated film of α-quinque-thiophene (α-5T) and P13. The two materials have been selected because of their transport and luminescence properties. α-5T is known as hole-transporting material,[20] and from pure reference devices we extracted a hole mobility of 2.5×10^{-2} cm^2/V s. P13 belongs to a class of perylene derivatives, which are well-studied electron-transporting materials. The pure reference device had an electron mobility of 5×10^{-3} cm^2/V s. The electron and hole mobilities of the pure materials lie within one order of magnitude, which is a prerequisite for achieving ambipolar current characteristics in a coevaporated film.

Exciton formation, and therefore light emission, strongly depend on the relative positions of the energy levels of the highest occupied (HOMO) and the lowest unoccupied molecular orbital (LUMO) of the two organic semiconductors. The HOMO level of α-5T lies at about −5.3 eV (Ref. 21) and the LUMO level at about −2.8 eV.[22] The values for the relevant energy levels of P13 were estimated from Ref. 23 as −5.4 eV and −3.4 eV for HOMO and LUMO, respectively. The molecular structures and the schematic device architecture are shown in Fig. 1. A heavily doped, *n*-type Si wafer (doping level: $10^{18}/cm^3$) with an aluminum back contact acts

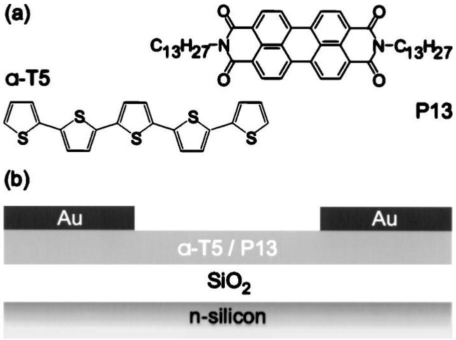

FIG. 1. (a) Molecular structure of α-5T and P13. (b) Device structure of the LEFET consisting of a coevaporated thin film of α-5T and P13.

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as gate electrode and substrate. The gate insulator consists of a thermally grown SiO_2 layer with a thickness of 150 nm. Prior to processing, the oxidized wafer was cleaned with a standard wet-cleaning procedure, comprising ultrasonic cleaning in acetone and isopropanol. The organic thin film (50 nm) used in this study was prepared by coevaporation of α-5T and P13 with a ratio of 1:1. The base pressure in the system was 2×10^{-7} mbar. The deposition rate was 0.3 Å/s for the coevaporated film. The Au source and drain contacts were thermally evaporated and had a thickness of 40 nm. The lateral dimensions were defined by shadow mask. The channel length and width of the coevaporated OFET were 40 μm and 55 mm, respectively. For reference experiments, single-layer OFETs of these two materials were fabricated with a channel length and width of 150 μm and 1200 μm, respectively. Au top contacts were used for both single-layer transistors. For characterization, the devices were placed in an argon glove box (<1 ppm O_2, H_2O). The transistor output and transfer characteristics were measured with a probe station using an Agilent 4155C semiconductor parameter analyzer. Simultaneously, the electroluminescence (EL) intensity was measured using a Hamamatsu S1336 photodiode. The charge-carrier mobility in ambipolar devices was extracted from the transfer characteristics, similar as for unipolar devices, using the saturated drain current $I_{D,\text{sat}}$ vs. V_G relation,[24]

$$I_{D,\text{sat}} = \frac{W}{L}\mu C(V_G - V_T)^2. \tag{1}$$

Here, W is the channel width, L is the channel length, μ is the charge-carrier mobility, C is the gate-oxide capacitance per unit area, V_G is the gate voltage, and V_T is the threshold voltage. To extract both the electron mobility and the hole mobility, it is necessary to measure the transfer characteristics for the negative as well as for the positive bias regime.

Figure 2(a) shows the output characteristics of a transistor with a coevaporated thin film of α-5T and P13. The figure is composed of two independent measurements applying either negative or positive gate and drain source voltage. Applying a negative gate bias V_G, typical p-channel characteristics are observed in the third quadrant for negative drain–source voltages with $|V_{DS}|\leq|V_G|$. With increasing $|V_{DS}|$, an abrupt, steep increase in the drain current I_D is measured, which is a typical characteristic of ambipolar operation in OFETs (see also Refs. 15, 16, and 19). This current increase is attributed to the injection of electrons into the organic thin film at the drain contact. A similar behavior is observed for positive gate bias in the first quadrant. The most striking feature, however, is the light emission monitored by the photocurrent of the photodiode, as shown in Fig. 2(b). For negative drain–source and gate voltages, the light output is apparently correlated to the nonsaturating drain current. The highest brightness is achieved for V_G=0 V and V_{DS} $=-50$ V. For positive drain–source voltages, only weak emission is observed. In contrast to the negative-voltage case, the emission here occurs at high gate voltages. The insert of Fig. 2(b) shows the light intensity as a function of I_D: For negative I_D the light output is proportional to the drain current. For positive voltages, on the other hand, the light output is independent of the drain current. To verify that light emission indeed originates in the recombination of electrons and holes injected by the drain and source electrodes, the gate current has been measured simultaneously. Even though a small gate leakage current was observed, no correlation between light emission and gate current could be found. The discontinuity in the current–voltage characteristics at 0 V drain–source voltage originates from the gate leakage at large gate bias. The magnitude of the gate leakage is the same for positive and negative voltages. However, he sign of the gate leakage changes with the sign of the gate bias.

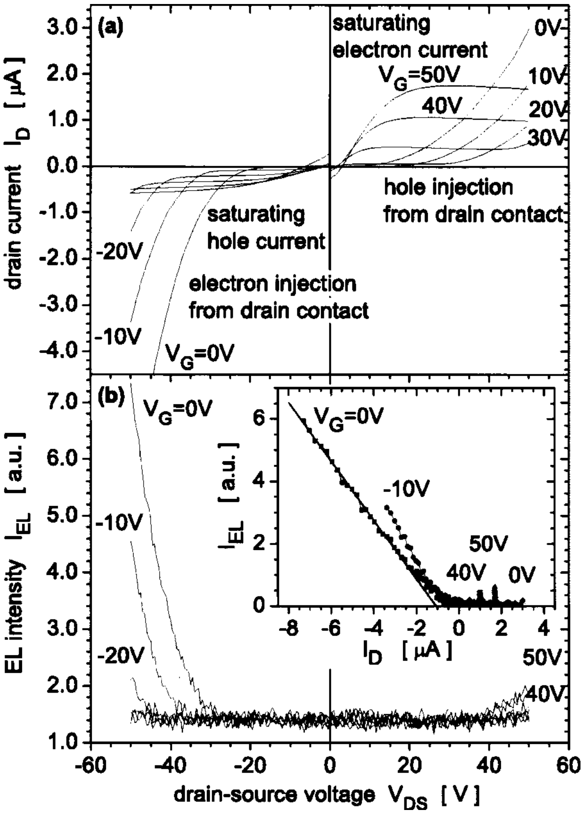

FIG. 2. (a) Output characteristics and (b) light intensity for the coevaporated α-5T/P13 thin-film transistor for negative and positive gate bias. The inset shows the photocurrent vs drain current for various gate voltages.

The light output from an ambipolar device is proportional to the recombination rate of electrons and holes between source and drain electrode. Assuming Langevin recombination,[25] the EL intensity I_{EL} is

$$I_{\text{EL}} \propto \int_0^L \frac{e}{\epsilon_r\epsilon_0}\cdot(\mu_n(x)+\mu_p(x))\cdot n(x)\cdot p(x)\cdot dx, \tag{2}$$

with ε_r the dielectric constant of the coevaporated organic layer, $n(x)$ and $p(x)$ the electron and hole densities, and $\mu_n(x)$ and $\mu_p(x)$ the electron and hole mobilities along the channel, respectively. Whereas the drain current I_D is a superposition of the electron and the hole current, the light intensity is determined by the $n(x)p(x)$ product. Therefore, no simple correlation of drain current and EL intensity seems to exist. A quantitative description of the ambipolar drain current, the hole and electron densities along the channel, and the light output will be given elsewhere.[26]

Figure 3 shows the transfer characteristics of the device. The figure is composed of two independent measurements applying either negative or positive gate and drain–source voltage. For large $|V_G|$, the current originates either from holes for negative values of V_G or from electrons for positive

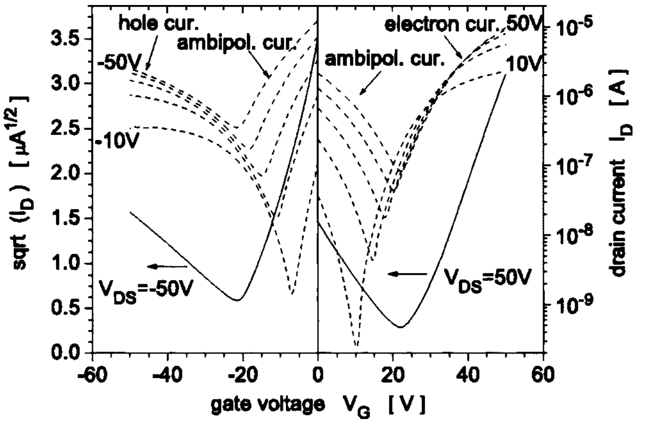

FIG. 3. Transfer characteristics of the coevaporated α-5T/P13 thin-film transistor for negative and positive gate bias. The solid lines show the square root of the drain current, the dashed lines the logarithmic drain–current vs gate voltage for various drain–source voltages V_{DS}.

values of V_G. The square root of the drain current I_D shows the expected linear dependence on V_G, as is known from unipolar devices. Contrary to unipolar devices, where typically a continuous increase in drain current $|I_D|$ is observed for absolute increasing gate voltage $|V_G|$, we observe first a decrease in $|I_D|$ for small values of $|V_G|$, which only starts to increase again after a certain value of $|V_G|$. This current originates in the corresponding opposite type of charge carrier. For increasing drain–source voltages the minimum in drain current shifts towards larger gate voltages. From the linear slope of the square root of I_{DS} versus V_G, a hole mobility of 10^{-4} cm^2/V s and an electron mobility of 10^{-3} cm^2/V s can be extracted. Compared with single-layer devices, the hole mobility in α-5T is two orders of magnitude smaller, whereas the electron mobility in P13 compares well with the one in single-layer devices. The influence of the coevaporation conditions on the mobility of each type of charge carrier will be investigated in more detail in the future. Again, we observe a discontinuity in the current–voltage characteristics at 0 V gate voltage. The ambipolar current for small negative gate voltages is carried primarily by electrons, for small positive gate voltage primarily by holes. Owing to the different mobilities of both carriers, the resulting drain currents have different magnitudes.

To summarize, we have demonstrated a light-emitting OFET based on a coevaporated thin film of α-5T and P13 that exhibits pronounced ambipolar conduction over a wide range of bias conditions accompanied by light emission. Light emission is correlated with the drain current, and can be modulated by both the drain–source voltage and the gate voltage. The device serves as an excellent model structure for a light-emitting OFET and demonstrates that by coevaporation of two different organic semiconductors, an electron- and a hole-transport material, the electron and hole mobilities can be adjusted, i.e., the resulting ambipolar characteristics are controlled by the stoichiometry of the two materials.

This work is funded within the EU-IST-FET program under project IST-33057 (ILO). The authors thank D. J. Gundlach (ETH Zurich), Th. Linder and G. Paasch (IFW Dresden) for useful discussions, and M. Melucci and G. Barbarella (CNR-ISOF Bologna) for providing α-5T. The authors acknowledge M. Tschudy, U. Drechsler, H. Riel, T. Beierlein (IBM ZRL), and R. Zamboni (CNR-ISMN Bologna) for support.

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