

Organic light emitting complementary inverters

Ebinazar B. Namdas,^{1,a)} Ifor D. W. Samuel,^{1,2,a)} Deepak Shukla,³ Dianne M. Meyer,³ Yanming Sun,¹ Ben B. Y. Hsu,¹ Daniel Moses,¹ and Alan J. Heeger^{1,a)}

¹Center for Polymers and Organic Solids, University of California, Santa Barbara, California 93106, USA

²Organic Semiconductor Centre, SUPA, School of Physics and Astronomy, University of St Andrews, North Haugh, St Andrews, Fife KY16 9SS, United Kingdom

³Kodak Research Laboratories, Eastman Kodak Co., Rochester, New York 14650, USA

(Received 20 August 2009; accepted 5 December 2009; published online 26 January 2010)

We show that p- and n-type light emitting field-effect transistors (LEFETs) can be made using “superyellow” as a light-emitting polymer, poly(2,5-bis(3-tetradecylthiophen-2-yl)thieno[3,2-b]thiophene) as a p-type material and a naphthalene di-imide as an n-type material. By connecting two of these LEFETs, we have demonstrated a light emitting complementary inverter (LECI). The LECI exhibited electrical and optical characteristics in the first and third quadrant of the transfer characteristics with voltage gain of 6 and 8, respectively. © 2010 American Institute of Physics. [doi:10.1063/1.3293293]

In recent years, organic semiconductors have attracted considerable attention because of their potential for creating low-cost portable electronic and optoelectronic devices such as organic light emitting diodes (OLEDs) and organic field-effect transistors (OTFTs).^{1–4} OLED performance and reliability are now sufficient for commercial products. Devices combining two or more functions are attractive because such integration gives not only an opportunity to simplify the design and fabrication processes but also leads to new applications. For example, organic light emitting field-effect transistors (LEFETs)^{5–12} combine the emission properties of an OLED with the switching properties of a transistor in a single device structure. This leads to new applications such as simplified pixels in flat panel displays and optoelectronic devices in communications, as well as having the potential for electrically driven lasers.¹³

Integration of organic p-type and n-type transistors in the same device architecture enables a complementary inverter to be made.^{14–19} This approach simplifies the design and fabrication of logic circuits (such as NOR, NAND, and ring oscillators). Several methods have been proposed to demonstrate inverters using structures that transport both electrons and holes; e.g., single layer ambipolar/bipolar materials,^{16,17} bilayer films,¹⁸ and blends of two components¹⁹ (p- and n-type organic semiconductors). In this letter, we demonstrate a light emitting complementary inverter (LECI). The LECI consists of two integrated LEFETs. This device combines the emission properties of a LEFET with the electrical properties of an inverter in a single device structure and thereby functions as an electrically driven optoelectronic switch. The electrical and optical characteristics of the LEFET and LECI are presented.

Figure 1 shows the device architecture and the molecular structures of the materials used. A heavily doped n-type silicon wafer functioned as the substrate and as the gate electrode. The n⁺⁺ Si gate electrode was first coated with 200 nm of SiO₂. A solution of poly(2,5-bis(3-tetradecylthiophen-2-yl)thieno[3,2-b]thiophene), PBTTT, (0.3% in chloroben-

zene), was then spin-coated onto the substrate at 3000 rpm to serve as a hole transporting layer. The samples were annealed at 150 °C for 10 min. in a N₂ glove box. The soluble PPV derivative, “superyellow” (SY), was spin-coated from solution at 2000 rpm to form the electroluminescent semiconducting polymer layer. A thin (20 nm) electron transporting layer of N,N'-bis(cyclohexyl) naphthalene-1,4,5,8-bis(dicarboximide),²⁰ (NDI), was vapor deposited on the top of the SY. The samples were then mounted onto a silicon shadow mask to complete the device fabrication using Au as source and drain electrodes. The channel length between

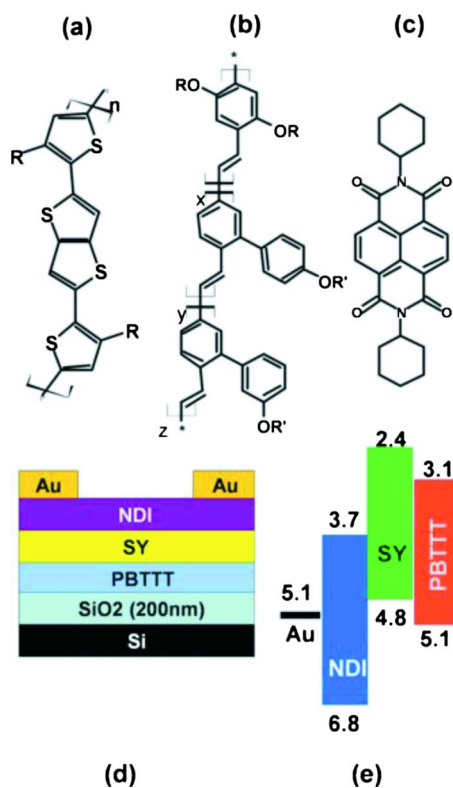


FIG. 1. (Color online) Molecular structure of (a) PBTTT; (b) SY; and (c) NDI layer; (d) Schematic diagram of the device architecture, and (e) energy level diagram showing HOMO and LUMO of NDI, SY, and PBTTT. The work function of Au is also shown.

^{a)} Authors to whom correspondence should be addressed. Electronic addresses: ebnamdas@ipos.ucsb.edu, idws@st-andrews.ac.uk, and ajhe@physics.ucsb.edu.

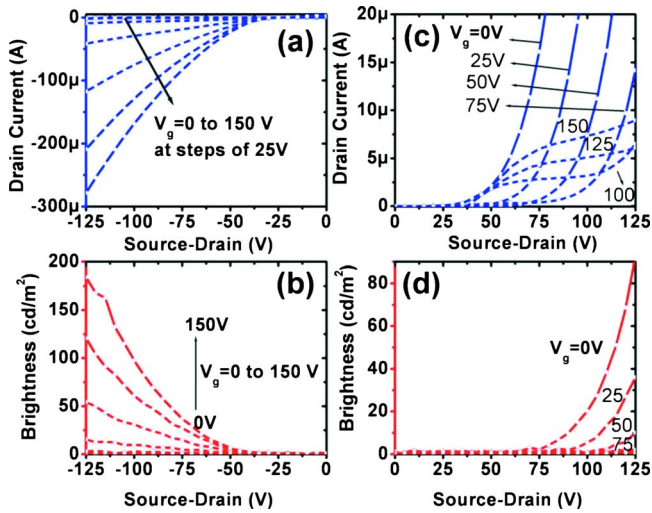


FIG. 2. (Color online) Electrical output (a) and optical output (b) characteristic of p-type voltage bias LEFET at various gate voltages. Electrical output (c) and optical output (d) characteristic of n-type voltage bias LEFET at various gate voltages.

source and drain electrodes was $19 \mu\text{m}$ and the channel width was $1000 \mu\text{m}$. All electrical and optical measurements were carried out in nitrogen atmosphere at <1.0 ppm oxygen; more details are presented in our previous publications.^{7,8}

Figures 2(a) and 2(c) show the electrical output characteristics of the LEFET in p- and n-bias corresponding to negative and positive gate voltages respectively. In p-type voltage bias, the transistor is biased negatively and the drain current was modulated with the applied gate voltage. The device demonstrates linear regimes for the drain current for lower source-drain voltages. A deviation from ideal saturation current behavior at higher source-drain voltages is observed. A possible reason for this could be the presence of interface charges at the NDI/SY interface.

In n-type voltage bias, the transistor is biased positively and the drain current (I_{DS}) increased superlinearly with increasing source-drain voltages (V_{DS}) at low values (0–75 V) of gate voltage. The I_{DS} decreases with increasing gate voltage and reaches a minimum value. Further increase in the gate voltage (100–150 V) leads to accumulation of electrons near the SY/NDI interface and transistor operates fully in n-channel mode. The gate leakage current at high voltage bias (150) was lower than the source-drain current by a factor of ~ 50 . These characteristics are typical for the ambipolar (or bipolar) LEFET. A deviation from ideal saturation current behavior at higher source-drain voltages is again attributed to the presence of interface charges at the NDI/SY interface.

The p- and n-channel field effect mobility μ_p and μ_n are estimated to be $\sim 5 \times 10^{-2}$ and $\sim 7 \times 10^{-3} \text{ cm}^2/\text{V s}$, respectively. The electron mobility of NDI is much lower than the previously reported mobility measured using OTS treated SiO_2 as the gate dielectric ($1\text{--}6 \text{ cm}^2/\text{V s}$).²⁰ Several factors may contribute to the much lower mobility for NDI on SY including, morphology, roughness of the film, and interface traps. In particular there is the possibility of a charge-transfer interaction at the NDI/SY interface which would result in charge trapping and explain the deviation from ideal saturation current behavior at higher source-drain voltages.

Bright yellow-green light emission was visible to the eye during the device operation in both p- and n-type voltage biases. The light emission was stationary and confined to a narrow region of $\sim 3 \mu\text{m}$ adjacent to the edge of the drain electrode. Figures 2(b) and 2(d) show the brightness, in units of cd/m^2 versus V_{DS} for p-type and n-type biases, respectively. The maximum brightness reached approximately $\sim 175 \text{ cd}/\text{m}^2$ for the p-channel. The data indicate excellent optical modulation by the gate voltage for the p-channel bias. For n-type voltage bias, the brightness increased super-linearly with increasing V_{DS} in the gate voltage range of 0–75 V and reaches a maximum brightness of $75 \text{ cd}/\text{m}^2$ at zero gate voltage. No light emission is observed for higher gate voltages.

We next consider the light emission and charge transport operating mechanisms in the LEFET. The energy level diagrams are shown in inset of Fig. 1. For p-type bias, the injection probability for the holes (from the Au electrode) into SY [highest occupied molecular orbital (HOMO) = 4.8 eV] and subsequently into PBTBT (HOMO = 5.1 eV) is much higher than that of electrons leading to the formation of a channel of accumulated holes near the gate insulator/PBTBT interface and the transistor operates in a p-channel accumulation mode. Since, the NDI is a good electron transporting material [lowest unoccupied molecular orbital (LUMO) = $\sim 3.7 \text{ eV}$], and is directly in contact with the Au electrode, there is relatively weak injection (or tunneling) of electrons into SY (LUMO = 2.4 eV). Thus, the holes moving across the channel recombine with electrons into the SY layer leading to the electroluminescent emission near the edge of the drain electrode.

For n-type bias [Fig. 2(d)] and at low gate voltage and higher V_{DS} , both electrons and holes are injected from source and drain electrodes into the SY and PBTBT layers. This leads to bipolar transport of electrons and holes at the interface of the SY/NDI and dielectric PBTBT, respectively, resulting in light emission near the edge of drain electrode. At much higher positive gate voltage (75–150 V), the probability of hole injection into SY and the PBTBT layer diminishes. As a result, there is no electroluminescence at higher gate voltage bias.

The maximum external quantum efficiency of the device was $\sim 8 \times 10^{-4}\%$. This value is much lower compared to our previously reported LEFET fabricated using SY.^{7,8} We consider that there are two major factors limiting the efficiency of our device. The first is poor charge capture to form an exciton, due to negative polarons being concentrated in the NDI and positive polarons in the SY. The other major factor is the strong offset in LUMO energy levels. This will lead to the dissociation of excitons formed near the NDI/SY interface, or prevent their formation in the first place.

In the following paragraphs, we discuss the fabrication and operation of the LECI. Inset of Fig. 3(b) displays a schematic diagram of the LECI that was fabricated by connecting two identical interdigitated LEFETs. The channel length and channel width of each LEFET were $50 \mu\text{m}$ and 3.45 mm , respectively. The gate is common to both LEFETs and serves as input node (V_{IN}). With the supply voltage V_{DD} and V_{IN} biased positively, a good inversion of the input signal at the output is observed as shown in the plot of V_{OUT} versus V_{IN} . As V_{IN} increases from 0 to 150 V at $V_{\text{DD}} = 150 \text{ V}$, V_{OUT} switches from high to low voltage, corresponding to switch-

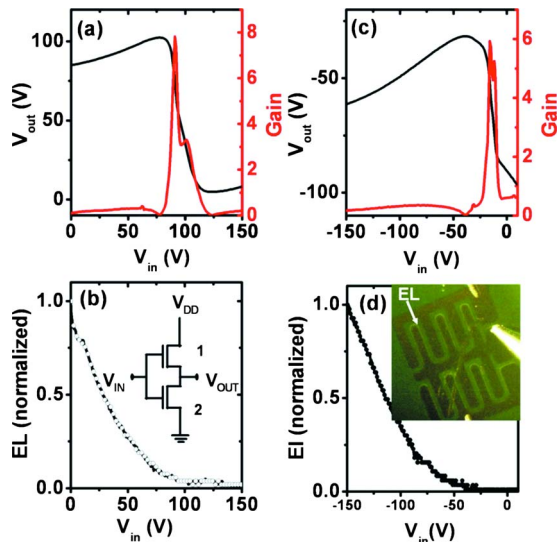


FIG. 3. (Color online) Electrical (a) and optical (b) characteristics of LECI operating in the first quadrant of transfer characteristics at $V_{DD}=+150$ V. Inset show schematic representation of the LECI; Electrical (c) and optical (d) characteristics of LECI operating in the third quadrant of transfer characteristics at $V_{DD}=-150$ V. Inset show optical microscope image of the LECI.

ing from a logic state of “1” to “0.” A gain of ~ 8 was obtained at nearly the half of the supply voltage. A close inspection of the device showed light emission in both transistors as V_{IN} was varied from 0 to 80 V. At $V_{IN}=0$ V, the light emission was the highest and found to decrease as V_{IN} increased. At $V_{IN} \geq 80$ V, no visible light was observed. These data demonstrate an electrically driven optoelectronic switch in which both optical and electrical high and low states corresponds to basic logic states of “1” to “0.” If the input and the bias voltage are negative, the device still operates as an optoelectronic switch with voltage gain of ~ 6 .

To understand the optoelectronic properties of the LECI, we first consider positive biases [Figs. 3(a) and 3(b)]. Under these conditions LEFET 1 acts like the p-type device of a regular complementary metal-oxide semiconductor inverter while LEFET 2 operates like the n-type device. When V_{IN} is high (80 to 150 V) or when $V_{IN}=V_{DD}$ [Fig. 3(a)], the gate to source voltage of n-type LEFET 2 equals V_{DD} which turns it ON. At the same time the gate to source voltage of p-type LEFET 1 is 0 V so it is OFF. The output node is pulled down to ground potential by current through the conducting n-type transistor. In this condition no light was visible in either LEFET. This can be explained by the fact that p-type LEFET 1 is OFF state, meaning no current flow in the device and hence no electroluminescence. For the n-type LEFET 2, the gate to source voltage is very high which, as mentioned earlier, is a situation for which there is no light emission.

On the other hand when V_{IN} is low (0 to 80 V) or when $V_{IN}=0$, the n-LEFET 2 is OFF since gate to source voltage is 0 V. The gate to source voltage of p-LEFET 1 is however equal to $-V_{DD}$, which turns it ON. In this condition light was visible in both LEFETs because p-type LEFET 1 is ON, resulting in electroluminescence from this transistor. For the n-type LEFET 2 the gate to source voltage is 0 (or low), and this causes n-type LEFET to operate in the bipolar transport

regime in which electrons and holes are transported simultaneously resulting in electroluminescence in the device as discussed in the previous paragraphs [Fig. 2(d)]. If the input and the bias voltage are negative [Figs. 3(c) and 3(d)], the inverter still operates with gain of ~ 6 , and the n- and p-channel function are exchanged between the two devices.

In summary, we have demonstrated p- and n-type LEFET operation with multilayer films that function as electron and hole transporting layers. The LEFET exhibited electrical and optical modulation at an applied gate voltage with brightness exceeding 175 cd/m^2 . We have demonstrated a LECI by connecting two identical LEFETs. The LECI exhibited electrical and optical characteristics in the first and third quadrants of the transfer characteristics with gain around 6–8. This multifunctional electrically driven optoelectronic switch could be useful in logic circuits as well as in communication devices.

Support for research was provided by the Air Force Office of Scientific Research (Charles Lee, Program Officer), the National Science Foundation (Polymer program, Grant No. NSF-DMR-0602280). A portion of this work was done in the UCSB nanofabrication facility, part of the NSF funded NNIN network. IDWS is an EPSRC Senior Research Fellow. The super yellow and PBTTT material was provided by Merck.

- ¹C. W. Tang and S. A. Vanslyke, *Appl. Phys. Lett.* **51**, 913 (1987).
- ²C. D. Dimitrakopoulos and P. R. L. Malenfant, *Adv. Mater.* **14**, 99 (2002).
- ³A. Dodabalapur, Z. Bao, A. Makhija, J. G. Laquindanum, V. R. Raju, Y. Feng, H. E. Katz, and J. Rogers, *Appl. Phys. Lett.* **73**, 142 (1998).
- ⁴J. D. Yuen, A. S. Dhoot, E. B. Namdas, N. E. Coates, M. Heeney, I. McCulloch, D. Moses, and A. J. Heeger, *J. Am. Chem. Soc.* **129**, 14367 (2007).
- ⁵J. Zaumseil, R. H. Friend, and H. Sirringhaus, *Nature Mater.* **5**, 69 (2006).
- ⁶J. S. Swensen, C. Soci, and A. J. Heeger, *Appl. Phys. Lett.* **87**, 253511 (2005).
- ⁷E. B. Namdas, P. Ledochowitsch, J. D. Yuen, D. Moses, and A. J. Heeger, *Appl. Phys. Lett.* **92**, 183304 (2008).
- ⁸E. B. Namdas, J. S. Swensen, P. Ledochowitsch, J. D. Yuen, D. Moses, and A. J. Heeger, *Adv. Mater.* **20**, 1321 (2008).
- ⁹M. Muccini, *Nature Mater.* **5**, 605 (2006).
- ¹⁰T. Rabe, M. Hoping, D. Schneider, E. Becker, H. H. Johannes, W. Kowalsky, T. Weimann, J. Wang, P. Hinze, B. S. Nehls, U. Scherf, T. Farrell, and T. Riedl, *Adv. Funct. Mater.* **15**, 1188 (2005).
- ¹¹P. Görrn, T. Rabe, T. Riedl, W. Kowalsky, F. Galbrecht, and U. Scherf, *Appl. Phys. Lett.* **89**, 161113 (2006).
- ¹²H. Nakanotani, S. Akiyama, D. Ohnishi, M. Moriwake, M. Yahiro, T. Yoshihara, S. Tobita, and C. Adachi, *Adv. Funct. Mater.* **17**, 2328 (2007).
- ¹³E. B. Namdas, M. Tong, P. Ledochowitsch, S. R. Mednick, J. D. Yuen, D. Moses, and A. J. Heeger, *Adv. Mater.* **21**, 799 (2009).
- ¹⁴H. E. Katz, A. J. Lovinger, J. Johnson, C. Kloc, T. Siegrist, W. Li, Y.-Y. Lin, and A. Dodabalapur, *Nature (London)* **404**, 478 (2000).
- ¹⁵S. De Vusser, S. Steudel, K. Myny, J. Genoe, and P. Heremans, *Appl. Phys. Lett.* **88**, 162116 (2006).
- ¹⁶Th. B. Singh, P. Senkarabacac, N. S. Sariciftci, A. Tanda, C. Lackner, R. Hagelauer, and G. Horowitz, *Appl. Phys. Lett.* **89**, 033512 (2006).
- ¹⁷T. D. Anthopoulos, D. M. De Leeuw, E. Cantatore, S. Setayesh, E. J. Meijer, C. Tanase, J. C. Hummelen, and P. W. M. Blom, *Appl. Phys. Lett.* **85**, 4205 (2004).
- ¹⁸Y. Sakamoto, T. Suzuki, M. Kobayashi, Y. Gao, Y. Fukai, Y. Inoue, F. Sato, and S. Tokito, *J. Am. Chem. Soc.* **126**, 8138 (2004).
- ¹⁹E. J. Meijer, D. M. De Leeuw, S. Setayesh, E. Van Veenendaal, B.-H. Huisman, P. W. M. Blom, J.-C. Hummelen, U. Scherf, and T. M. Klapwijk, *Nature Mater.* **2**, 678 (2003).
- ²⁰D. Shukla, S. F. Nelson, D. C. Freeman, M. Rajeswaran, W. G. Ahearn, D. M. Meyer, and J. T. Carey, *Chem. Mater.* **20**, 7486 (2008).