AIP Applied Physics Letters

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Citation: Applied Physics Letters **93**, 223301 (2008); doi: 10.1063/1.3027057
View online: http://dx.doi.org/10.1063/1.3027057
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/93/22?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS **93**, 223301 (2008)

Light-emitting polymer space-charge-limited transistor

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(Received 19 June 2008; accepted 25 October 2008; published online 1 December 2008)

Polymer light-emitting transistor is realized by vertically stacking a top-emitting polymer light-emitting diode on a polymer space-charge-limited transistor. The transistor modulates the current flow of the light-emitting diode by the metal-grid base voltage. The active semiconductor of the transistor is poly(3-hexylthiophene). Yellow poly(*para*-phenylene vinylene) derivative is used as the yellow emitting material. As the cathode is fixed at -12 V and the grid base voltage varies from 0.9 to -0.9 V the light emission is turned on and off with on luminance up to 1208 $\mathrm{cd/m^2}$. The current efficiency of the light-emitting transistor is 10 cd/A. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3027057]

Semiconductor devices based on conjugated polymers have been widely explored for polymer light-emitting diode (PLED), field-effect transistor (FET), and solar cell. One of the promising applications of the polymer devices is the thin, flexible, and lightweight active-matrix display.[1] Unlike liquid-crystal display, which is voltage-controlled, in polymer display a PLED in each pixel is controlled by a driving transistor, which supplies the current. The current output of the transistor must be very stable and uniform. Currently polycrystalline silicon FET suffers from a uniformity problem, while amorphous silicon and organic FET have rather low carrier mobility and long-term stability problems.[2] In addition, FET is a horizontal device while PLED is a vertical device, so they can only be placed side by side and the aperture ratio of the pixel is limited by the size of the FET. One possible way to replace the conventional combination of LED driven by FET is the organic light-emitting transistor, which combines the electrical switching and the light generation capabilities in a single device.[3] However, so far the applications of light-emitting transistor is hindered because the emission efficiency is rather low, and the emission zone is in a narrow line pattern, which is unfavorable for display applications. In this work, a non-FET polymer light-emitting transistor is demonstrated by vertically integrating a top-emitting PLED on a polymer vertical metal-base transistor. The vertical transistor is used to modulate the current and therefore the luminance of the PLED. Uniform luminance over 1000 $\mathrm{cd/m^2}$ is achieved. The current efficiency of the light-emitting transistor is 10 cd/A at yellow emission. The aperture ratio is basically 100% because the light emitted upward is not shielded by the vertical metal-base transistor underneath with roughly the same area. Light generation in vertical transistor was reported before,[4,5] but these early works show poor luminance no more than 1 $\mathrm{cd/m^2}$. The high luminance is achieved by a vertical polymer light-emitting transistor, the space-charge-limited transistor (SCLT). This transistor has a high output current density around 27 $\mathrm{mA/cm^2}$, which can drive PLED to high luminance.

Organic vertical metal-base transistors with various architectures and operation principles have been developed[6–16] to overcome the limits of conventional horizontal organic FETs. The channel length of vertical metal-base transistor is defined by the thickness of the organic layer, and the output current is modulated by a base electrode embedded in the organic semiconductor. Two types of vertical metal-base transistors, namely the hot-carrier transistor[13,14] and SCLT,[15,16] show promising performance. In this work the polymer SCLT is vertically integrated with a PLED. The carriers are injected from the emitter into the semiconductor, passing through the openings on the metal grid base, and finally arriving at the collector. The potential distribution and current flow between emitter and collector is controlled by the voltages of grid. Because the emitter-collector distance is only tens of nanometers, high current output and high speed can be achieved even for low carrier mobility typical for PLED, thus solving the problems of organic FETs for current output and speed. The fabrication is even simpler than FET as no photolithography is needed. In addition the high current output and potentially high speed SCLT shows far better stability than the organic FET. Due to the current channel confinement, organic FET characteristics are easily affected by the chemical and physical defects at the gate dielectric-semiconductor interface. SCLT is free from the unpredictable conditions at the dielectric interface because its current is uniformly distributed in the bulklike organic light-emitting diodes.

The device structure of the polymer SCLT with a silicon monoxide insulating layer on the aluminum grid is shown in Fig. 1(a). A 400 Å layer of poly(3,4-ethylenedioxythiophene) doped with polystyrene sulfonated acid (PEDOT:PSS) is spin-coated onto the indium tin oxide (ITO) glass substrate to flatten the ITO surface and serve as the emitter for holes. A layer of 200 Å poly(3-hexylthiophene) (P3HT) is then spin coated from chlorobenzene solution (1.5 wt %). After annealing at 200 °C for 10 min, a spin rinsing with xylene is used to remove the remaining soluble part of P3HT. The substrate is submerged into 2000 Å (1000 Å) polystyrene spheres ethanol solution with 0.24 wt % (0.4 wt %) for 40 s and then transferred into a beaker with boiling isopropanol solution for 10 s. The substrate is immediately blown dry in a unidirectional nitrogen flow to form two dimensional colloidal arrays without aggregation. After 150 Å Al grid is evaporated as grid; the sample is annealed in high vacuum (10^{-6} torr). 500 Å SiO is evapo-

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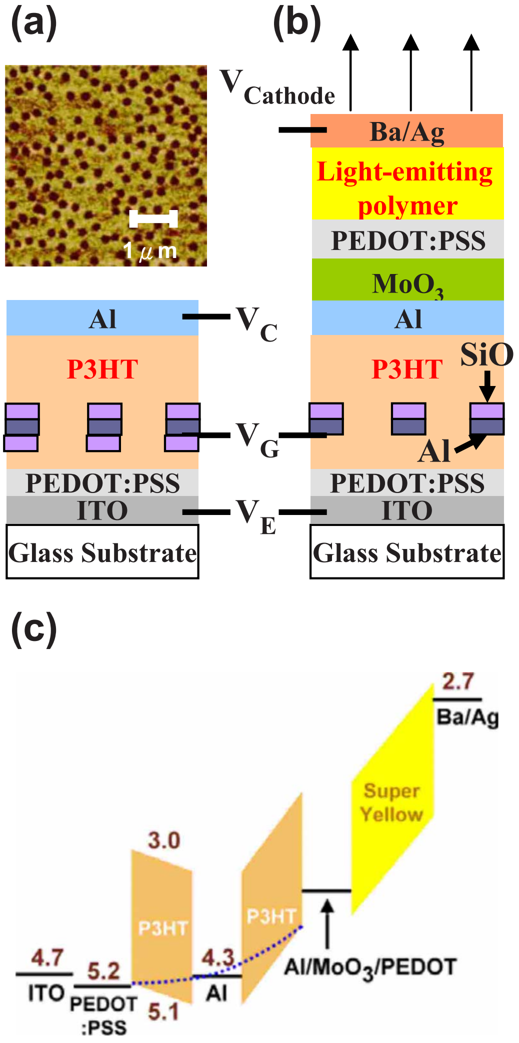

FIG. 1. (Color online) (a) Device structure of the polymer SCLT and the AFM image of Al grid with 2000 Å opening diameter. (b) Device structure of the top-emitting polymer light-emitting transistor. (c) Energy profile of the top-emitting polymer light-emitting transistor.

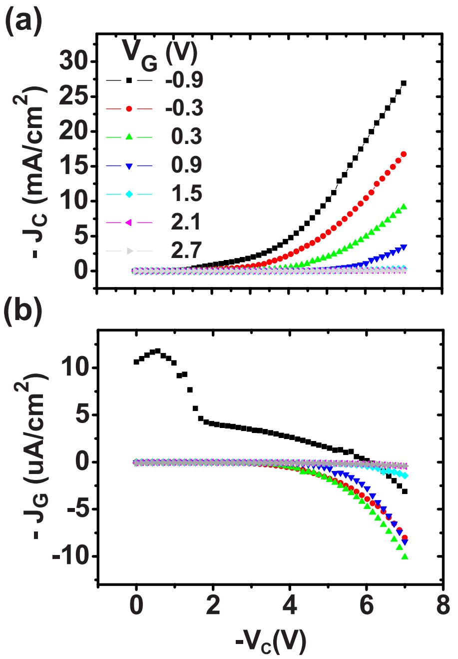

FIG. 2. (Color online) The electric characteristics of the polymer SCLT with 2000 Å openings on Al grid. (a) Collector current density J_C and (b) grid current density J_G as a function of collector voltage V_C at various grid biases V_G.

rated to prevent large leakage current from grid to collector. After the polystyrene spheres are removed by an adhesive tape, another layer of 800 Å P3HT is spin coated from xylene solution (3 wt %). A 400 Å Al collector is finally deposited to complete the polymer SCLT with active area of 3 mm^2. The atomic force microscope (AFM) image of Al grid with 2000 Å opening diameter is also shown in Fig. 1(a), which is taken after removing the polystyrene spheres. The opening density is about 7 openings/μm^2. For the light-emitting transistor as shown in Figs. 1(b) and 1(c), the fabrication procedure of the vertical transistor is the same as described above, while the procedures of the PLED are described below. A 30 Å MoO_3 anode is deposited on the Al collector with same shadow mask, followed by spin coating 450 Å PEDOT:PSS from an aqueous solution diluted by isopropanol in equal volume. The diluted PEDOT:PSS solution contains also 2 wt % surfactant (Zonyl FSN) to facilitate aqueous PEDOT:PSS solution to be spinned on a substrate coated with hydrophobic polymer. Poly(*para*-phenylene vinylene) polymer derivative (Super Yellow by Merck) is spin coated from toluene as light-emitting layer of 100 nm. The semitransparent cathode Ba(100 Å)/Ag(150 Å) is then deposited to complete the device with an active area of 4 mm^2. Due to convenience in mask alignment the LED area is slightly larger than the transistor area of 3 mm^2. The PEDOT:PSS layer prevents the transistor from being dissolved by the solution of the light-emitting polymer.

The polymer SCLT is in a diode sandwich structure with a third metal base electrode inserted into the semiconductor to control the vertical carrier flow between the emitter and the collector. In this work, additional high vacuum annealing after Al grid deposition is used for increasing the P3HT mobility and current density of ITO/PEDOT:PSS/P3HT/Al diode. The transistor output current can be enhanced from 1 mA/cm^2 to as high as 27 mA/cm^2. The improved polymer SCLT with grid opening diameters of 2000 Å is shown in Fig. 2(a). J_C and V_C are the collector current density and the collector voltage, respectively. The ITO electrode is commonly grounded and the Al collector is negatively biased at V_C. Various grid voltages are applied to Al grid base to modulate the current. The negative collector current density J_C means the holes flow from the transistor and are collected by the Al collector. An insulating SiO layer is deposited on Al grid to reduce the grid leakage current from Al grid to collector. Such SiO layer allows a higher collector bias and higher output current before grid diode breaks down. When V_C is -7 V and V_G is -0.9 V, the transistor output current density is about 27 mA/cm^2 and the on/off ratio is 386. The maximum on/off ratio is 428 when V_C is reduced to -6 V. J_G as a function of V_C is also shown in Fig. 2(b). The remaining J_G may come from the leakage path in the nondense SiO layer and the Al sidewall close to the opening. As the transistor is on, the current gain J_C/J_G of this transistor is around 10^4. Besides, the annealing procedure is expected to have two results. First, annealing in high vacuum causes oxygen dedoping of P3HT. Second, annealing helps the polymer chains to reorganize to have a high mobility and high current density. The collector current density of 27 mA/cm^2 is about the same as PLED under normal operation. The

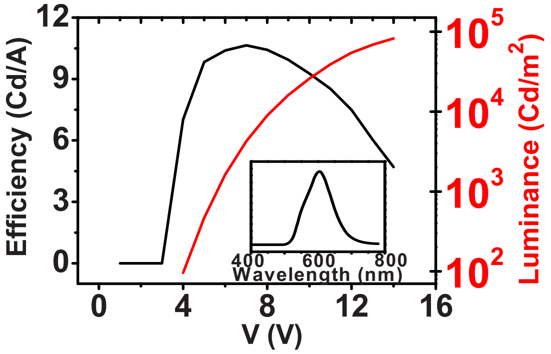

FIG. 3. (Color online) The efficiency and luminance of the top-emitting PLED.

transistor is therefore able to drive a PLED vertically stacked with roughly the same area.

The characteristics of the top-emitting Super Yellow PLED with the structure ITO/PEDOT:PSS/Super Yellow/Ba/Ag is given in Fig. 3. The current efficiency is 10.6 cd/A. In the light-emitting transistor the PLED is fabricated on top of the Al collector of SCLT with a MoO_3 layer in between. The electrical characteristics of the polymer light-emitting transistor with opening diameters of 1000 and 2000 Å on Al grid are shown in Figs. 4(a) and 4(b), respectively. This structure is effectively a PLED connected in series to a variable resistor, which modulates the PLED current and therefore luminance. The ITO emitter electrode is commonly grounded and various V_G is applied to the Al grid base. The voltage applied to the semitransparent cathode V_{cathode} is varied from 0 to −12 V.

Voltage V_C of the Al collector in the SCLT component is floating and determined by the voltage drop across the PLED. It spontaneously matches the currents of the SCLT component and the PLED component. The luminance at various V_G for V_{cathode} at −12 V is shown in Fig. 4. The maximum luminance around 500 and 1200 cd/m^2 is achieved for polymer light-emitting transistor with grid opening diameters of 1000 and 2000 Å, respectively. The minimum luminance can be turned off by increasing V_G until large leakage current occurs. This device not only fulfills the minimum luminance requirement for the display panel, but also achieves by far the highest luminance among various light-emitting transistor principles. The on-off contrast of the luminance is yet to be improved. The residual off-luminance cannot be switched off by the grid voltage before grid diode breakdown. A stronger insulation of the grid is needed to withstand a more positive grid voltage.

The structure and operation principle of the polymer light-emitting transistor above is different from previous report on static induction transistor,[5] where the light is emitted inside the channel of the vertical transistor. In our device the light emission and driving are separated into different components of the device with a floating electrode between them. The performance of each component is therefore not compromised by the integration with the other component. The composite electrode Al/MoO_3/PEDOT:PSS layer has two advantages. First, the electrons injected from the cathode will not reach P3HT. The efficiency and color of the light emission are therefore not ruined by the unwanted recombination in P3HT. Second, the electrons injected from the cathode will not arrive at the Al grid and cause a large J_G and poor current gain.

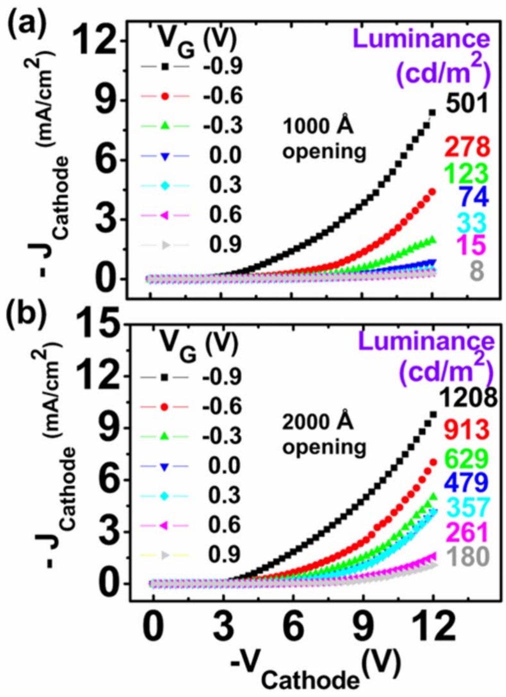

FIG. 4. (Color online) The electric characteristics of the polymer light-emitting transistor with (a) 1000 Å and (b) 2000 Å openings on Al grid. The luminances at $V_{\text{Cathode}}=-12$ V are also demonstrated for various V_G condition.

In summary, the performance of the polymer SCLT is enhanced by annealing and by insulator on grid metal to have high output current density and high current gain. The top-emitting polymer light-emitting transistor based on the SCLT is demonstrated. This transistor has low voltage, high light-emission aperture ratio, and high luminance good enough for display applications.

This work is supported by the National Science Council of Taiwan under Contract No. NSC96-2112-M-009-036. The authors thank Merck, Inc. for providing the Super Yellow polymer.

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