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Vertical polymer phototransistor featuring photomultiplication due to base-field shielding

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We introduce a vertical polymer phototransistor with low operational voltage (-1.5 V). A blended polymer layer with both acceptor and donor materials was used as a channel material in the vertical space-charge-limited transistor. Under illumination, we obtained external quantum efficiency (EQE) as high as 360% at 620 nm. We propose the effects of base-field shielding as a means to explain high EQE. This proposition has been supported by two-dimensional simulation of the device. © *2011 American Institute of Physics*. [doi:10.1063/1.3552714]

Organic photodetector arrays can be fabricated at low temperatures on a large scale, with low production costs. The integration of organic photodetectors and other organic electronic devices such as organic transistors, organic light-emitting diodes, and various sensors enables the development of numerous devices, including footprint scanners, proximity sensors, and biomedical sensors.[1–3] A good photodetector requires high photoresponsivity, low operational voltage, wide bandwidth, and processes that are compatible with organic transistors, should one desire an active-matrix photodetection array. Conventional organic photodiode can usually be operated at low voltages (<5 V) with wide bandwidths (over 80 MHz).[4,5] Unfortunately, the processes involved in the fabrication of vertical structures are incompatible with the process of forming horizontal organic thin-film transistor.

In this letter, we propose a vertical organic photo transistor (OPT) based on vertical polymer transistors, space-charge-limited transistors (SCLTs).[6] This SCLT-based OPT utilizes a photodetecting mechanism referred to as the base-field shielding to induce photomultiplication (PM). To enhance the effect of base-field shielding, we exchanged the channel material from a donor material, poly(3-hexylthiophene) (P3HT), to a blend of a donor material and an acceptor material, P3HT blended with (6,6)-phenyl-C61-butyric acid methyl ester (PCBM), to enhance the dissociation of excitons.

The structure of the device is shown in Fig. 1(a). The dimensions of the structure are marked. The fabrication processes are similar to those described in our previous report.[6] Conditions different from those in previous report are addressed as follows. First, a 50-nm-thick SiO_x is evaporated following the base metal (a 40-nm-thick aluminum) evaporation to serve as the upper insulator of the grid structure. Second, the thicknesses of the PVP and P3HT were approximately three times greater than those of the previous report, thereby enabling greater control of the device current leakage. Third, P3HT or P3HT blended with 0.1 wt % PCBM was used as the polymer semiconducting material. A light-emitting diode backlight was used as the light source to irradiate the sample from the indium tin oxide (ITO) electrode.

In this study, P3HT-SCLT exhibited characteristics similar to those reported in our previous study.[6] A high on/off current ratio ($>10\,000$) was obtained at a low collector-to-emitter operational voltage (V_{CE}) (-2.2 V). The transfer characteristics (collector current density J_C as a function of V_{BE}) of P3HT-SCLT in the total darkness and under illumination when $V_{CE}=-1.5$ V are shown in Fig. 1(b). In the total darkness, the off-current was below 5×10^{-4} mA/cm^2. Under illumination, the off-current increased significantly. If the photocurrent density (J_{ph}) is defined as the current density under illumination (J_{ill}) minus the current density in total darkness (J_{dark}), J_{ph} of 1 mA/cm^2 is obtained when the illumination intensity is 11 mW/cm^2, $V_{BE}=1.5$ V, and $V_{CE}=-1.5$ V. We can assume that the high J_{ph} in P3HT-SCLT was not produced by the photogenerated carriers in P3HT
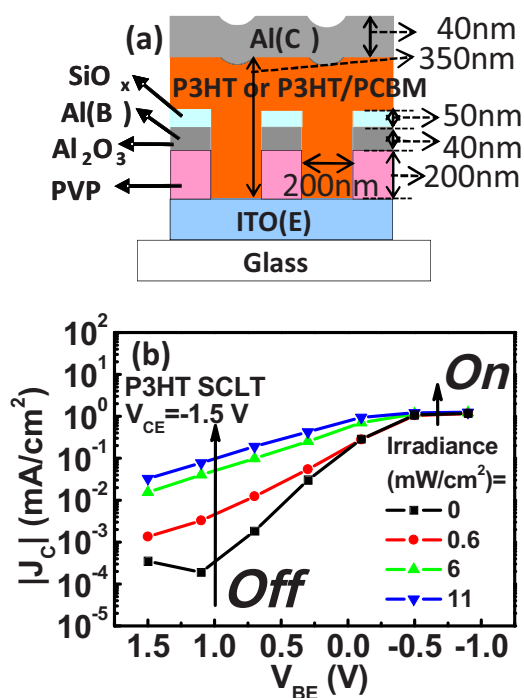

FIG. 1. (Color online) (a) Schematic illustration of the structure of P3HT or P3HT:PCBM-SCLT. (b) Transfer characteristics of a P3HT-SCLT in total darkness and under illumination.

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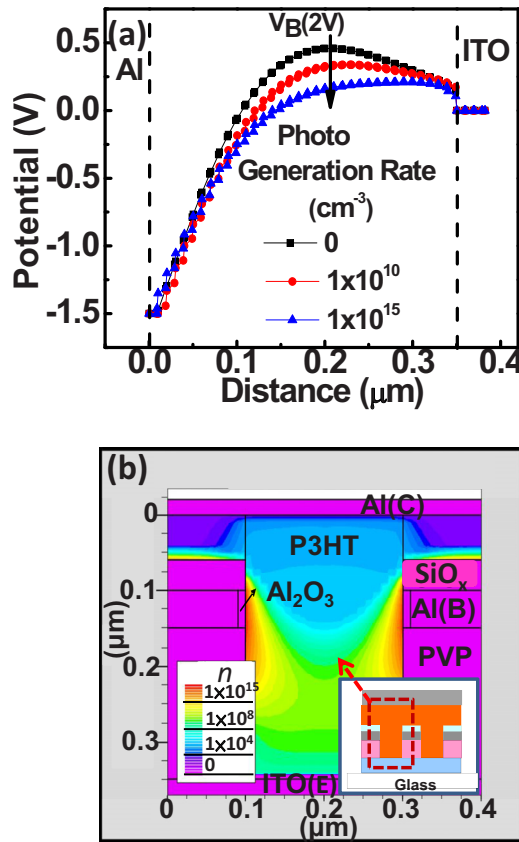
FIG. 2. (Color online) (a) A simulated potential distribution at the central vertical channel. (b) The two-dimensional electron distribution of P3HT-SCLT with a photogeneration rate $=10^{10}$ cm^{-3} and a $V_{BE}=2$ V. Electron concentration is denoted by n.
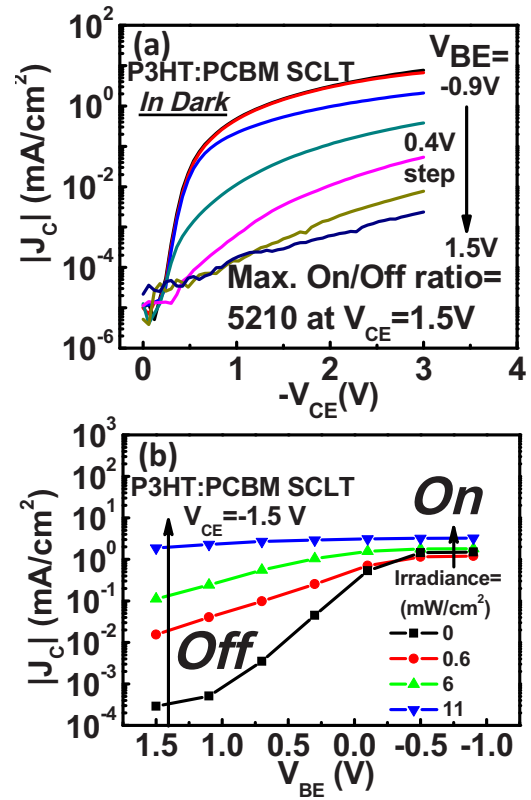

FIG. 3. (Color online) (a) Output characteristics of the P3HT:PCBM-SCLT. (b) Transfer characteristics of a P3HT:PCBM-SCLT in total darkness and under illumination.

due to the fact that exciton dissociation in P3HT without an acceptor blending is known to be poor. A low J_{ph} of 3 $\times 10^{-4}$ mA/cm^2 was obtained with an illumination intensity of 11 mW/cm^2 and $V_{CE}=1.5$ V.

To investigate the mechanism causing high J_{ph} in P3HT-SCLT, we simulated carrier distribution and the potential channel profile with TCAD SILVACO ATLAS software. The two-dimensional simulation was performed by setting $V_{CE}=-1.5$ V and $V_{BE}=2$ V to simulate the device operated in its off-state. Material parameters were defined as in Ref. 7. The potential distribution in the central channel from emitter to collector is shown in Fig. 2(a). In total darkness (i.e., photo-generation rate$=0$ cm^{-3}), the emitter was unable to inject holes into the channel because the 2 V V_{BE} created a potential barrier to impede hole transport, whereby a low off-state current was achieved. Under illumination, the potential barrier formed by the 2 V V_{BE} significantly dropped, and with a lower potential barrier, a high off-state current was expected. A lower potential barrier under illumination was explained by the two-dimensional electron distribution plot shown in Fig. 2(b). With a photogeneration rate$=10^{10}$ cm^{-3} and $V_{BE}=2$ V, the electrons generated by exciton dissociation accumulate around the base of the electrode. The accumulated electrons shield the base-field and suppressed the influence of base bias on the channel potential. As a result, the potential barrier caused by the base bias was lowered under illumination. The off-state current, with exponential dependence on the potential barrier height, increased significantly. However, due to poor exciton dissociation in P3HT, the concentration of electrons around the base electrode was not high

enough to completely shield the base-field. Therefore, in Fig. 1(b), we see that the off-state current density under irradiance of 11 mW/cm^2 is still 40 times lower than the on-state current density. If complete base-field shielding is expected for further enhancement of the photoresponse, the exciton dissociation in the channel region will need to be improved. In the following experiment, we added PCBM to P3HT to form a donor/acceptor interface to improve exciton dissociation.

The characteristics of the P3HT:PCBM-SCLT are shown in Fig. 3(a). When the base to emitter potential (V_{BE}) changed from -0.9 to 1.5 V, the device was switched from on- to off-state. A maximum on/off ratio of approximately 5000 was obtained at a collector to emitter potential (V_{CE}) of -1.5 V. The on/off ratio is inferior to that of P3HT-SCLT because electron injection from the collector metal to PCBM caused a leakage in the current. An electron blocking layer between the collector and active layer could be applied to reduce the leakage in future studies. In this work, to maintain a reasonable on/off ratio, the P3HT:PCBM blending ratio was kept to 1:0.1 by weight. If the P3HT:PCBM blending ratio were 1:0.5 or 1:1, the device would suffer from large leakage current, which would degrade switching performance. The transfer characteristics of a P3HT:PCBM-SCLT in total darkness and under illumination are shown in Fig. 3(b). V_{CE} is fixed as -1.5 V. J_C in total darkness exhibits significant on-state and off-state when V_{BE} changes from negative to positive. Under illumination, both on-state J_C and off-state J_C increased with an increase in the intensity of illumination. The on/off ratio decreased with increases in the intensity of illumination, indicating a weaker base control over the channel. When the light intensity was 11 mW/cm^2
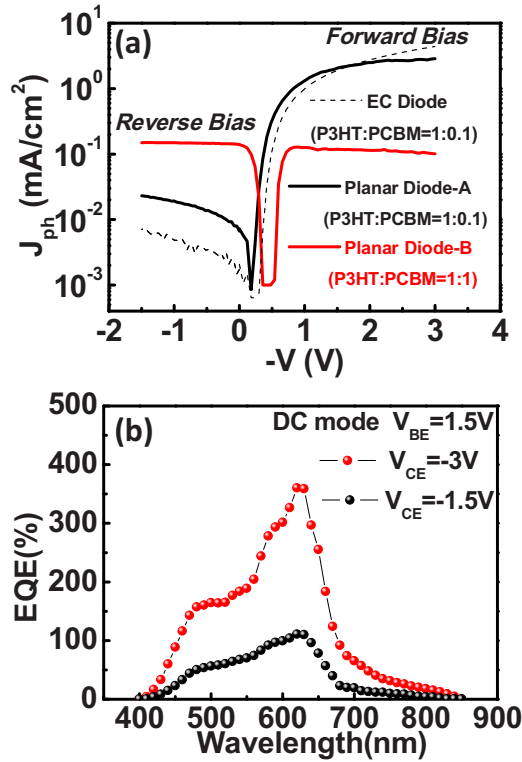
FIG. 4. (Color online) (a) J_{ph} of planar diode of P3HT:PCBM=1:0.1 (diode-A), planar diode of P3HT:PCBM=1:1 (diode-B), and EC diode in P3HT:PCBM-SCLT (P3HT:PCBM=1:0.1) plotted as a function of bias voltage. (b) The EQE of P3HT:PCBM-SCLT.

the off-state J_C was nearly equal to the on-state J_C. The base-field was completely shielded and V_{BE} lost control over J_C.

In Fig. 3(b), it is interesting to note that the on-current also increases significantly under illumination. Because the SCLT operated in on-state is similar to a forward-biased diode, we needed to analyze the photoresponse of a P3HT:PCBM diode in both reverse- and forward-biased conditions. We fabricated two planar ITO/P3HT:PCBM/Al diodes: one with a P3HT:PCBM blending ratio of 1:0.1 (diode-A) and the other with a P3HT:PCBM blending ratio of 1:1 (diode-B). J_{ph} (i.e., $J_{ill}-J_{dark}$) of these two diodes was plotted as a function of bias voltage in Fig. 4(a), wherein the intensity of illumination was 11 mW/cm^2. In reverse-biased conditions, diode-A had a much lower J_{ph} than diode-B, because the low PCBM percentage suppressed exciton dissociation. In forward-biased conditions, J_{ph} of diode-A was 20 times higher than that of diode-B. Obviously, exciton dissociation does not explain the large J_{ph} in forward-biased conditions. At equal dissociation, the photocurrent is reported to be different depending on the bias conditions.[8] We propose that the hole injection from ITO to P3HT is enhanced under illumination. In previous reports, photoinduced carrier injection was a result of carrier trapping at the injection interface to lower the injection barrier.[9] J_{ph} of the emitter to collector (EC) diode in P3HT:PCBM-SCLT is shown by a dotted line in Fig. 4(a). We obtained a J_{ph} higher than 1 mA/cm^2, when the forward-biased voltage was 1.5 V (i.e., $V_{CE}=-1.5$ V). Although the forward-biased diode-A and the forward-biased EC diode had a high J_{ph}, their photocurrent-to-dark current ratios $[J_{ph}/J_{dark}=(J_{ill}-J_{dark})/J_{dark}]$ were smaller than 1 be-

cause of the high J_{dark}. In P3HT:PCBM-SCLT, the current flowing through a forward-biased EC diode (i.e., $V_{CE}=-1.5$ V) was turned off by the base potential in total darkness and, as a result, a low J_{dark} was achieved. Under illumination, base electrodes lose control over channel current due to the effect of base-field shielding. At the same time, the effects of photoenhanced hole injection in the forward-biased EC diode contribute to high channel current. The photocurrent-to-dark current ratios (J_{ph}/J_{dark}) were 4×10^2, 4×10^3, and 6×10^3 when illumination intensities were 0.6, 6, and 11 mW/cm^2, respectively.

Finally, we measured the EQE of the P3HT:PCBM-SCLT with an active area of 1 mm^2, $V_{CE}=-1.5$ V, and $V_{BE}=1.5$ or 3 V, as shown in Fig. 4(b). In SCLT, when irradiated from an ITO electrode, only a fraction of the active area comprised P3HT:PCBM material. The EQE measured in this case was actually an effective EQE per unit active area. The maximum EQEs of P3HT:PCBM-SCLT were 110% and 360% at 620 nm when $V_{BE}=1.5$ and 3 V, respectively. An EQE higher than 100, representing a PM phenomenon, was dominated by a base-field shielding effect.

In summary, we demonstrated a vertical polymer phototransistor, based on the SCLT. The photoresponse in SCLT was governed by the following steps. First, the SCLT was operated in the off-state to provide a current in total darkness. The EC diode was forward biased, and the base potential created a potential barrier to impede hole transport. Second, under illumination, excitons were generated in the polymer channel layer. In this study, the channel material was P3HT blended with PCBM at a blending ratio of 1:0.1 to enhance exciton dissociation. Third, after exciton dissociation, electrons flowed toward the base electrode. Electrons accumulated around the base electrode shield of the base-field. As a result, the potential barrier was reduced. At the same time, the hole injection from ITO into the channel is enhanced. Finally, with the reduction in the potential barrier and the enhancement of hole injection, a large photocurrent was obtained.

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[7]The thicknesses of PVP, Al grid, SiO$_x$, and P3HT are 200, 40, 50, and 350 nm, respectively. The opening diameter is 200 nm. The highest occupied molecular orbital and lowest unoccupied molecular orbital levels of P3HT are 5.2 and 3.0 eV. The work functions of emitter and collector are 5.2 and 4.3 eV. The hole mobility and electron mobility in P3HT are 10^{-5} and 10^{-6} cm^2/V s.

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