Organic inverter circuits employing ambipolar pentacene field-effect transistors

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Ambipolar transport has been observed in pentacene films grown on polyvinyl alcohol gate dielectric with hole and electron mobilities of 0.3 and 0.04 $cm^2/V\,s$, respectively. A simple device structure with Au as source-drain electrode can be used to operate a transistor in both *p*-channel and *n*-channel modes without employing low work function metal electrodes for ambipolar charge injection. Using ambipolar pentacene field-effect transistors, we construct a complementarylike inverter with voltage inversion gain of ~10. These inverters are able to operate both in first and third quadrants of the voltage output to voltage input characteristics which is a unique feature of employing ambipolar transistors. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2235947]

Ambipolar charge transport, the simultaneous or selectively variable transport of electrons and holes in organic field-effect transistors, is of interest from both fundamental science and from applications point of view.[1–3] Such devices capable of transporting both electrons and holes have additional advantage in comparison to devices which transport single types of charge carriers (unipolar). A unique example of application of use of ambipolar organic field effect transistors (OFETs) is a light emitting organic field-effect transistors (LEOFETs).[4] Ambipolar OFETs would allow balanced charge carrier injection of holes and electrons placing the recombination zone between source-drain electrodes to be tuned by gate voltage, hence improving the quantum efficiency. In comparison, for unipolar devices, light emission is restricted to a region very close to the contacts which injects the charge carriers of the lower mobility.

Ambipolar transistors employing soluble derivatives of methanofullerene as an active layer have been utilized to demonstrate complementarylike inverters.[5,6] These inverters are able to operate in both (first and third) quadrants of transfer characteristics giving a unique opportunity for achieving high noise margin. In contrast, inverters with unipolar transistors operate only in one quadrant (i.e., either in the first or third).[7,8] Crone *et al.* achieved complementary inverter circuits by separating the transistor channels in order to facilitate the separate vacuum deposition of the two semiconductors (a *n* and a *p* type), thus making circuit fabrication complex.[9] While preparing this letter, Ahles *et al.*[10] published their work where an inverter circuit based on interface doped pentacene layer has been demonstrated. This approach to some extent simplifies the device fabrication by utilizing a single semiconductor layer. However, two separate source-drain electrodes adjacent to each other have to be prepared one with high work function and another low work function electrode to achieve ambipolar operation. Moreover, Ahles *et al.*[10] have observed no *n*-channel operation of pentacene OFET by employing high work function (Au) source drain electrodes. In general low work function electrodes are not suitable for efficient hole injection due to large energy barrier between electrode work function and highest occupied molecular orbital (HOMO) level of the semiconductor. Most ambipolar OFETs reported to date show relatively low charge carrier mobilities.[1–3,5,11,12] For example, in the work by Yasuda *et al.*[12] the maximum ambipolar charge carrier mobilities are in the order of 10^{-5} $cm^2/V\,s$ (for electrons) and 10^{-4} $cm^2/V\,s$ (for holes) of the pentacene OFETs using Ca as source-drain electrodes. For high operating frequency of complementary organic circuits, ideally, one would like to have OFETs with high carrier mobilities, smaller dimensions, and with much simpler device configuration.

We have recently discovered that with a proper choice of different surface energy organic dielectrics, pentacene OFETs can exhibit ambipolar operation even when a high work function metal (Au) is employed as the source and drain electrodes.[13] We found that the carrier mobilities in these pentacene devices are in the order of 0.3 $cm^2/V\,s$ for holes and 0.04 $cm^2/V\,s$ for electrons. The key improvement made to the ambipolar transistor since our previous report was to demonstrate an ambipolar inverter circuit with voltage inversion characteristic gain of ~10.

These devices as shown in Fig. 1(a) are fabricated on indium tin oxide (ITO) glasses which were cut into 15 $\times 15$ mm^2 size. Etching procedure was done leaving an ITO gate electrode area of 0.5 mm (width) $\times$ 15 mm (length) as a gate electrode. Polyvinyl alcohol (PVA) (Mowiol® 40-88) with an average molecular weight of 120 000 (Sigma-Aldrich) was used as received. PVA (10% in water) were spun at 1500 rpm on top of the ITO glass. We kept the polymer dielectric solution stirring for two days before spin coating to avoid any undissolved particles. After cleaning the ITO substrate with acetone, 2-propanol in ultrasonic bath polymer dielectric was spin cast from solution. To ensure that all the solvents were removed from the PVA film, these

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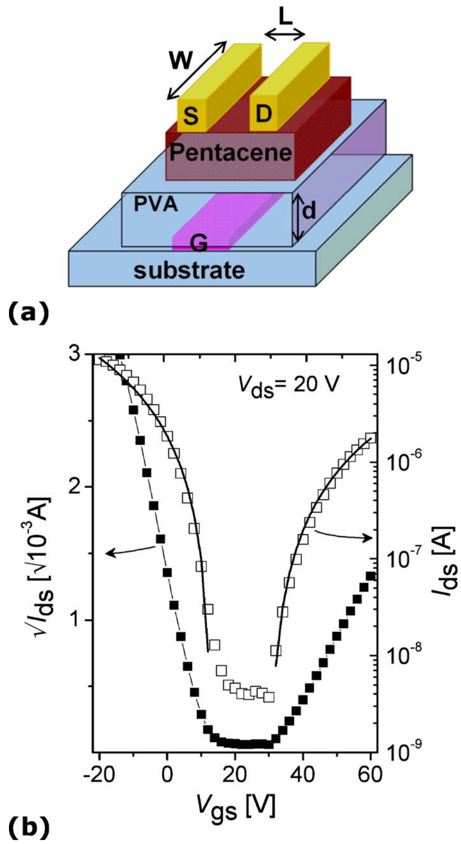

FIG. 1. (Color online) (a) Schematics of the top-contact ambipolar pentacene OFET device structure with Au as source (S)-drain (D) electrode with ITO as a gate (G) electrode. (b) Transfer characteristics of ambipolar pentacene OFET on PVA demonstrating electron-enhancement as well as hole-enhancement mode. $\sqrt{I_{ds}}$ vs V_{gs} plot (solid line) is also plotted. The lines are fitted to the data using Eq. (1).

dielectric films were dried in Ar atmosphere at 60 °C overnight. The average thickness of the dielectric layer was around 1 μm. Pentacene was used as received without further purification from Sigma-Aldrich. Organic thin films were grown at the rate of 0.2–0.3 Å/s at room temperature at a base vacuum of 10^{-6} mbar using Leybold Univex 350 system which have source to substrate height of about 0.5 m. Subsequently metal evaporations were done at a vacuum of 9×10^{-7} mbar using a shadow mask. Separate metal-insulator-semiconductor (MIS) structure devices with Au (60 nm) as top electrodes were prepared for the quasistatic capacitance-voltage measurements. Devices were transported from the organic evaporation chamber to the glovebox for metal evaporation and further all electrical characterization was carried out under an argon environment. The channel length L of the device is 25 μm and the channel width is W=1.4 mm, giving a W/L ratio of 56. An Agilent E5273A with two source-measure unit instrument was employed for the steady state current-voltage measurements. For dynamic response time measurement, we used a combination of function generator and voltage amplifier to give an ac voltage pulse and a scope to measure the input and output signals as depicted. For dc characteristics all measurements were performed with a scan rate of 2 V/s unless otherwise stated. The thickness of the dielectric film was measured in ambient condition with a Digital Instrument Dimension 3100 atomic force microscope. Comparison of measured dielectric capacitances in inert condition with the thickness of the dielectric layer consistently gave the values of capacitance C_i of 1.8 nF/cm^2.

Figure 1(b) depicts the transfer characteristics (source-drain current I_{ds} as a function of source-gate voltage V_{gs} for a fixed source-drain voltage V_{ds}) which demonstrate the ambipolar operation of pentacene FET with Au as a source-drain electrode. Figure 1(b) also shows $\sqrt{I_{ds}}$ vs V_{gs} which represents a sharp turn on of both p-channel and n-channel transistors with threshold voltages V_{th} of 13 and 30 V for hole and electron-enhanced modes, respectively. It is to be noted that we have used a rather thick dielectric layer ($\approx$1 μm) on the expense of large V_{gs}. I_{ds} depends quadratically on the applied V_{gs}. This allows us to estimate the mobility μ from well known transistor equation:

$$I_{ds} = \frac{\mu W C_i}{2L}(V_{gs} - V_t)^2, \tag{1}$$

where W is the channel width, L is the channel length, and C_i is the dielectric capacitance. With a channel length of 25 μm, width of 1.4 mm (W/L ratio of 56), and dielectric capacitance of 1.8 nF/cm^2, with drain currents of 11 μA (hole current at V_{gs}=−20 V) and 1.7 μA (electron current at V_{gs}=60 V), saturated mobilities of 0.3 and 0.04 cm^2 V/s is estimated for holes and electrons, respectively. We note that the electron mobility reported here is comparable with that of interface doped pentacene devices.[10] The most dramatic difference between the results reported previously for an interface doped pentacene and the present undoped pentacene film grown on organic dielectric is the use of pairs of calcium and gold source and drain electrodes to obtain high electron and hole mobility for the former case. In our estimation of mobility, we used standard transistor equation (1) without taking into account for contact resistant[14] and assumed V_{gs} independent mobility for simplicity. However, we also observed (not shown here) a similar V_{gs} dependence of μ with that of pentacene films grown on self-assembled monolayered alumina dielectric.[15] V_{gs} dependence of μ is explained using various models[15] which attributed to the defect induced charge traps in the semiconductor film. Our morphology study reveals a comparatively uniform and smaller grain size when pentacene films are grown on PVA dielectric.[13]

The recent demonstration of complementarylike voltage inverter circuit based on soluble derivatives of fullerene based ambipolar FETs (Ref. 5) has prompted us to investigate the complementarylike inverters using ambipolar pentacene FETs. A typical transfer characteristic of the ambipolar pentacene inverter is shown in Fig. 2 along with the scheme of the complementarylike voltage inverter circuit employed using the ambipolar pentacene FETs. Two identical ambipolar pentacene OFETs are connected in such a way that both of the gates have a common input node (V_{in}) as depicted in the inset of Fig. 2(a). With the supply voltage, V_{dd} and V_{in} are biased positively, a good inversion of the input signal at the output is observed as shown in the plot of V_{out} vs V_{in} with a characteristic voltage gain of ~12 in the first quadrant. We note that transfer characteristics presented in Fig. 2(a) presents unique features of negligible pull up or pull down of V_{out} which is normally observed in other complementarylike inverters based on other organic semiconductors.[9] We have also observed that the inverter operates in the third quadrant of the V_{in} vs V_{out} plot, as shown in Fig. 2(b). Similarly a

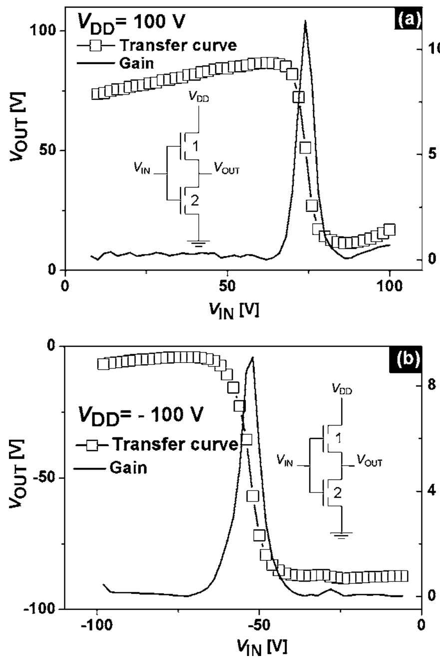

FIG. 2. Transfer characteristics of complementarylike inverter using two identical ambipolar pentacene OFETs. (a) Inverter characteristics (solid line) with V_{in} and V_{dd} being positively biased and the corresponding gain. The inset shows the inverter circuit configuration. (b) Inverter characteristics with V_{in} and V_{dd} being negatively biased. Inset shows the scheme of the inverter circuit employed.

characteristic gain of ~9 is observed here. We note here that this is the specific advantage of having ambipolar transistor for an inverter to be able to operate in both quadrants. The dynamic response of the inverters is depicted in Fig. 3(a). Dynamic response measurement as shown in Fig. 3(b) was performed by measuring the output signal V_{out} with respect to the input signal V_{in} using an oscilloscope with 1 MΩ input

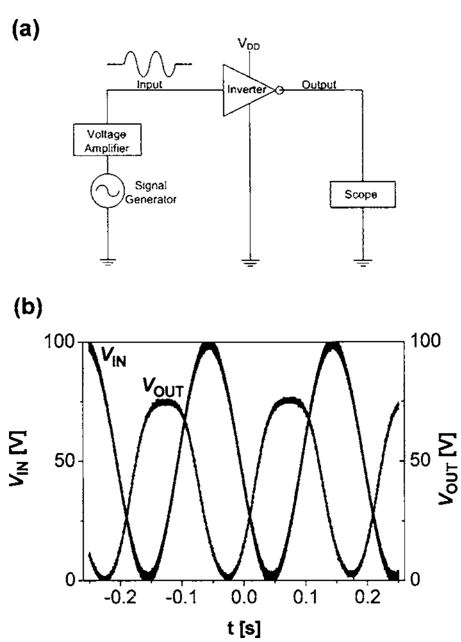

FIG. 3. (a) Schematic diagram of the experimental setup used for the measurement of oscillating frequency of the complementarylike voltage inverters shown in the inset of Fig. 2. The output and input signals are measured simultaneously using a digital oscilloscope with an input impedance of 1 MΩ. (b) Oscillating signal measured at the output of inverter. We note that the inverter oscillates also for negative V_{dd}.

impedance. Parasitic capacitance has to be minimized through the optimization of the geometry (gate electrode area, drain-source electrodes, and area of the semiconductor film as well as thickness of the dielectric). There is some controversy on the validity of using ambipolar devices to build a complementary circuit. The controversy stems from the fact that ambipolar transport prevents the device from switching off, because either electron or hole transport occurs at all gate biases. However, Fig. 1(b) actually shows that there is a lag between the threshold voltages for *n*- and *p*-channel regimes, thus leaving place for a gate voltage range where the device is indeed switched off.

The detailed reason for obtaining ambipolar transport with pentacene film grown on PVA dielectric is still under investigation. On the other hand, these results can also be a basis for obtaining ambipolar transport for many organic materials with high electroluminescence efficiency in the light of studies of light emitting transistors for balanced charge carrier injection.

In conclusion, high mobility ambipolar pentacene organic field-effect transistors have been demonstrated. Using such ambipolar OFETs, the complementarylike inverters have been demonstrated. These inverters are also able to operate in first and third quadrants of transfer characteristics with gains around 10.

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