AIP | Applied Physics Letters

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Citation: Applied Physics Letters **92**, 253307 (2008); doi: 10.1063/1.2939553
View online: http://dx.doi.org/10.1063/1.2939553
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/92/25?ver=pdfcov
Published by the AIP Publishing

Balancing the ambipolar conduction for pentacene thin film transistors through bifunctional electrodes

Chuan-Yi Yang,[1] Dhananjay,[2] Shiau-Shin Cheng,[1] Chun-Wei Ou,[1] You-Che Chuang,[1] Meng-Chyi Wu,[1] and Chih-Wei Chu[2,3,a)]
[1]*Institute of Electronics Engineering, National Tsing Hua University, Hsinchu 30013, Taiwan*
[2]*Research Center for Applied Sciences, Academia Sinica, Taipei 11529, Taiwan*
[3]*Department of Photonics, National Chiao-Tung University, Hsinchu 30010, Taiwan*

(Received 19 March 2008; accepted 14 May 2008; published online 27 June 2008)

We report an effective way to equilibrate hole and electron transport in ambipolar pentacene thin film transistors (TFTs). It was achieved by inserting an ultra thin lithium fluoride (LiF) layer at the electrode/organic interface to form a bifunctional source/drain electrode. It can be observed that the presence of ultrathin LiF layer will facilitate the injection of electrons from source/drain electrodes to organic semiconductor, resulting in a significant enhancement of the *n*-channel conduction, without sacrificing *p*-channel conduction for ambipolar pentacene TFTs. These factors make them potential candidates for the organic complementary circuits and inverter circuits of these ambipolar TFTs are also demonstrated. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2939553]

Organic logic circuits utilizing the complementary technology, with the advantages of high noise margin, low power dissipation, and robust operation, have attracted a great deal of attention from academic and industrial researchers.[1] Organic thin film transistors (OTFTs) with ambipolar transport are desirable for complementary technology and there are several methods reported in literature, to achieve ambipolar organic field effect transistors (OFETs) through bilayers,[2] blends,[3] and single-component materials.[4] However, those approaches reported to date, focus on the injection of both holes and electrons into organic semiconductors from the same metal electrodes. In such structures, poor carrier transport for at least one type of charge carriers originates from the mismatch of energy level between the electrodes and semiconductor. In order to overcome such an injection barrier for one of the carrier types, researchers have used an asymmetric device structure with high- and low-work function metals as source and drain (S/D) electrodes, respectively.[5] Unfortunately, this approach complicated the fabrication process and the fabricated complementary inverters may not be able to operate in both (first and third) quadrants for achieving high noise margin.

It has been reported that engineering of the organic/electrode interfacial properties is crucial for achieving the balanced hole and electron injection for high performance ambipolar operations.[6,7] Recently, there have been various attempts made to improve the electron injection from an Al cathode to an emitting layer by inserting a thin layer of alkali metal halides[8] and carboxylates.[9] Actually, those interlayer not only enhance the fraction of injected electrons, but also allow the top electrode to contact with the organic semiconductor, maintaining a symmetric device structure. Thus, in such a symmetric device structure, it is expected that both holes and electrons could be efficiently injected by inserting a thin interlayer. Although numerous research groups have inserted interlayer to enhance *n*-channel conduction for OTFTs,[10] no reports were conducted wherein they function as bifunctional electrodes for ambipolar transport in organic semiconductors. Henceforth, in this article, we demonstrate an effective way to balance hole and electron transport in ambipolar pentacene TFTs by inserting a LiF layer at the electrode/organic interface to serve as bifunctional S/D electrode.

The substrate of these ambipolar transistors is p^+-doped Si with thermally grown 300 nm SiO_2. The deposition parameters for the sol-gel coated polymethyl methacrylate thin films on the SiO_2 gate oxide, its role as a surface modifying layer and the pentacene layer deposition procedure are explained elsewhere.[11] Ultrathin LiF layer was then deposited using thermal evaporation onto the pentacene film and the thickness of the LiF layer was varied from 0 to 30 Å. Finally, 50 nm thick aluminum was thermally evaporated onto the pentacene film through a shadow mask to form the S/D electrodes. The thickness of the films was monitored by using a quartz crystal monitor. The typical channel length and width of the devices were 100 μm and 2 mm, respectively. The electrical measurements of the devices were performed in a nitrogen environment inside a glove box using HP

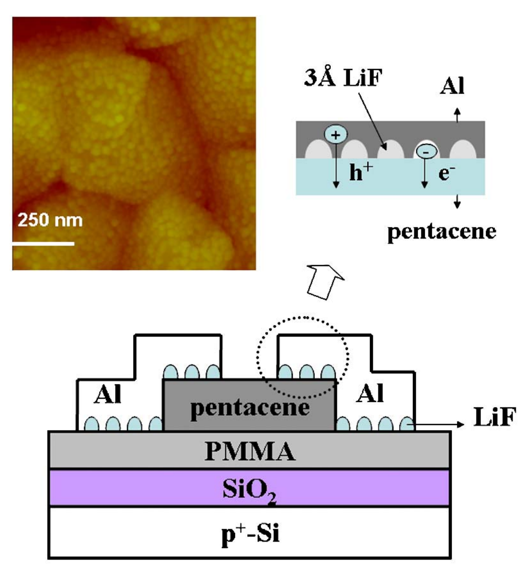

FIG. 1. (Color online) A schematic cross-sectional configuration of our top-contact OTFTs. The inset is the atomic force microscope image of p^+-Si/SiO_2/pentacene/LiF.

[a)]Electronic mail: gchu@gate.sinica.edu.tw.

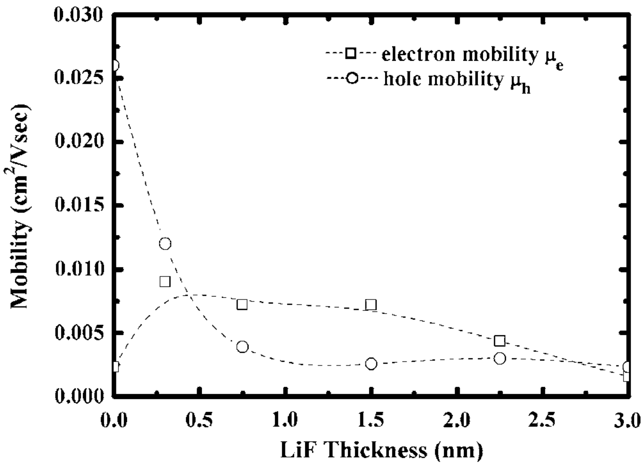

FIG. 2. Electron and hole mobilities as a function of LiF layer thickness for the ambipolar pentacene OFETs.

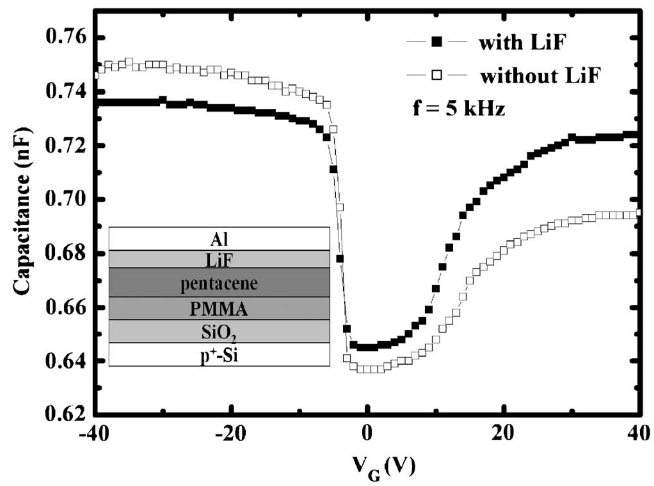

FIG. 3. Capacitance (C) as a function of gate voltage (V_G) for the ambipolar pentacene field-effect transistors with and without a LiF interlayer at 5 kHz.

4156C and Keithley 4200 semiconductor parameter analyzer. The capacitance-voltage (C-V) measurement was performed by Agilent E4980A precision *LCR* meter.

The schematic cross section of device structure used in this study is shown in Fig. 1. Since the range of thicknesses of LiF thin film employed lies in the subnanometer regime, the film surface presumed to be discrete with discontinuous islands [inset, Fig. 1]. This results in the formation of two different junctions at metal/organic semiconductor interface, which are pentacene/LiF/Al and pentacene/Al. The presence of discontinuous LiF islands leads to a lowering of the energy barrier for electrons from Al to the lowest unoccupied molecular orbital (LUMO) of pentacene resulting in an increase of electron injection via tunneling.[12] Meanwhile, holes can be readily injected to the highest occupied molecular orbital of pentacene from Al. Figure 2 shows the dependence of hole and electron mobilities on the LiF film thickness. As shown in Fig. 2, the device with bare Al as S/D electrodes exhibited unbalanced bipolar conduction, with the mobility of holes one order in magnitude higher than that of electrons. However, the effective contact area of the pentacene/LiF increase with an increase in LiF thickness, which in turn enhances the injection capability for electrons. As a result, the electron mobility is marginally enhanced and the *p*-type behavior is partially inhibited rather than totally sacrificed. Further increase in the LiF layer thickness beyond 3 nm leads to the diminishing of balanced bipolar conduction. This could be attributed to the high resistivity of the LiF layer, which leads to slow down the tunneling probability for electrons and to reduce the effective contact area of pentacene/Al. Therefore, the thicker LiF layer makes the electron and hole injections difficult for the ambipolar conduction.

Figure 3 shows the variation of capacitance (C) as a function of gate voltage (V_G) for the metal-insulator-semiconductor (MIS) structures with and without a LiF interlayer at 5 kHz. There are three distinct regions of accumulation, depletion, and inversion. It is well known that, in the OTFTs and organic light-emitting diodes (OLEDs) the injection of electrons mainly relies on the low-work function metals such as Al. Further enhancement of electron injection could be achieved by introducing the ultra-thin layers of LiF or Alq_3 mainly due to several mechanisms such as chemical reaction models and dipole models.[13] From the C-V measurements it was observed that both electrons and holes are accumulated at interface relying on the applied gate bias. As compared to the MIS structure with a pristine Al as the top electrode, the population of accumulated electrons is much lower than that of the device with a LiF interlayer. On the contrary, the accumulated holes of the MIS device with bare Al are slightly more than those of the device with a LiF interlayer. The accumulated hole and electron charges calculated from the area under the C-V curves for the devices with

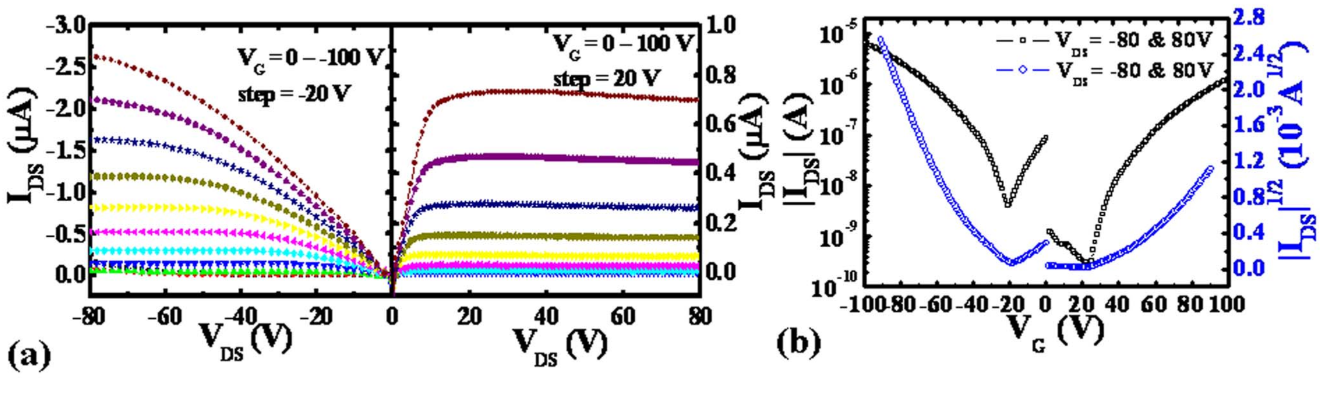

FIG. 4. (Color online) (a) Drain-source current (I_{DS}) as a function of drain-source voltage (V_{DS}) of ambipolar pentacene FETs with LiF (3 Å)/Al as bifunctional electrodes at various gate voltages in p- and n-channel operations. (b) The corresponding plots of $|I_{DS}|$ and $|I_{DS}|^{1/2}$ as a function of V_G for the ambipolar pentacene FETs with LiF (3 Å)/Al as bifunctional electrodes.

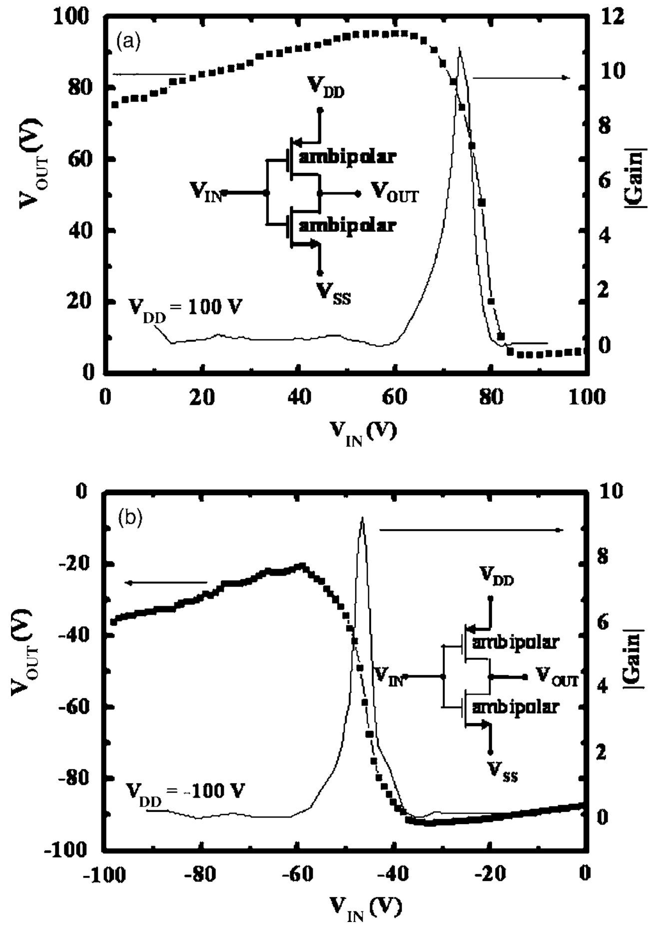

FIG. 5. Transfer characteristics of complementarylike ambipolar pentacene inverter in the (a) first and (b) third quadrants with their corresponding gains. The insets show the scheme of the complementarylike inverter circuit, which is composed of two identical pentacene transistors.

and without the LiF interlayer are 2.89×10^{-8} and 2.8×10^{-8} C and 2.95×10^{-8} and 2.68×10^{-8} C, respectively. These results illustrate the strategic importance of the discontinuous LiF film to enhance the electron injection, and thus to increase the capacitance at positive bias while preserving the excellent hole accumulation capability. The enhancement of the electron injection is probably attributed to the reduction in the potential barrier across the Fermi level of metal electrode and the LUMO of pentacene, which is possibly resulted from the ultrathin LiF layer.[14]

Figure 4(a) shows the drain-source current (I_{DS}) as a function of drain-source voltage (V_{DS}) of ambipolar pentacene FETs with LiF (3 Å)/Al as bifunctional electrodes at various gate voltages. The devices show an apparent ambipolar transport behavior at a gate voltage of larger than $|40|$ V. It can be noted that both the n- and p-channel output characteristics exhibit significant saturation which behaves quadratically as a function of gate bias. Figure 4(b) represents the corresponding plots of $|I_{DS}|$ and $|I_{DS}|^{1/2}$ as a function of V_G for the ambipolar pentacene FETs with LiF (3 Å)/Al as bifunctional electrodes. The threshold voltage (V_T) and carrier mobility can be extracted from the intercept and slope of the curve of $|I_{DS}|^{1/2}$ versus V_G. These devices exhibit strong field-effect modulations of channel conductance in the ambipolar operation with a threshold voltage of −17 and 39 V for p- and n-type operations, respectively. In addition, the mobility values for hole and electron are estimated as 1.2×10^{-2} and 9×10^{-3} cm^2/V s, respectively. Such a balanced electron and hole mobilities makes the feasibility of ambipolar transport in pentacene TFTs.

One of the most essential and important building blocks of a logic circuit is the inverter, and these pentacene-based OFETs mentioned above are suitable for the feasibility of an inverter. A complementarylike inverter circuit is shown in the inset of Fig. 5. Because of the unique ambipolar characteristics, the inverter is capable of operating in both the first and third quadrants. Figures 5(a) and 5(b) show the typical transfer characteristics of such an inverter. A sharp inversion is observed in both quadrants. When the supply voltage (V_{DD}) and input node (V_{IN}) are biased positively, the ambipolar transistors can be operated as p-channel and n-channel FETs, the inverter works in the first quadrant with the maximum voltage gain of 9.5; whereas the supply voltage (V_{DD}) and input node (V_{IN}) are biased negatively, the ambipolar transistors can be operated as n-channel and p-channel FETs, and the inverter works in the third quadrant with the maximum voltage gain of 11.5.

In summary, we have demonstrated that the insertion of a LiF interlayer between pentacene and Al electrode can significantly improve the performance of ambipolar pentacene TFTs. As compared to the OTFTs with pristine Al electrodes, the devices with an LiF interlayer exhibit an enhanced electron mobility without scarifying a great deal of hole mobility. Employing the discontinuous islands of LiF as an interlayer is an effective and simple method to fabricate high performance OTFTs for practical applications.

The authors are grateful to the National Science Council (NSC), Taiwan (95-2218-E-001-003) and the Thematic Project of Academia Sinica, Taiwan for financial support.

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