A light emitting transistor based on a hybrid metal oxide-organic semiconductor lateral heterostructure

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A light-emitting field-effect transistor was fabricated, with its architecture based on a distinct heterojunction located midway between the source and drain contacts. Tetracene enabled hole transport on one side of the heterojunction (hole mobility ~0.071 cm^2/Vs), while amorphous solution-processed zinc tin oxide supported electron transport on the other side (electron mobility ~0.81 cm^2/Vs). The drain current vs. gate voltage curves of this device have a bell-shaped profile that is characteristic of lateral heterojunction bipolar field-effect transistors. The green light emission—from tetracene—closely follows the trend in drain current and is naked-eye visible in a darkened room. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3689758]

Organic light-emitting field-effect transistors (OLEFETs) combine light emission and addressability into a single device and could, therefore, be an advantageous display element. The possibility of light emission from a field-effect transistor (FET) was initially discussed in Ref. 1, and a number of groups have succeeded in creating these devices, *albeit* approaching the device architecture in different ways. OLEFETs in the literature are FETs that are typically based on one of the following: (1) a single layer of pure, unipolar semiconductor,[2,3] (2) a single layer of ambipolar semiconductor (either as a pure material[4,5] or as a binary blend of a p-type with an n-type[6]), (3) a layered stack consisting of a p-type on top of or below an n-type[7] and possibly with an emitter layer sandwiched in between,[8] or (4) a lateral heterostructure that has a p-type and n-type each occupying roughly half the length of the channel and forming a heterojunction inside the channel.[9]

Lateral heterostructure light-emitting organic field-effect transistors (LH-LEFETs) are bipolar in nature, and under suitable drain and gate voltages, a hole channel and electron channel form in the p- and n-type semiconductors, respectively. These channels allow injected holes and electrons to drift from their respective electrodes to the heterojunction and recombine, with the possibility of light emission if the recombination process is radiative in one or both semiconductors. As such, at least one of the two semiconductors must be light-emissive. Also, both semiconductors should possess reasonably high and balanced charge carrier mobilities. Due to the additional patterning required to fabricate the LH-LEFET, mechanical and chemical robustness are highly advantageous.

In this manuscript, we report the electrical and light emission characteristics of an LH-LEFET based on the p-type organic semiconductor tetracene in combination with the n-type inorganic semiconductor zinc tin oxide (ZTO). Tetracene is a well-known electroluminescent semiconductor with high hole mobility and has been used in unipolar OLEFETs.[2] Among oxide semiconductors, ZTO is an amorphous semiconductor[10] that was chosen for a number of its attributes: first, its electron mobility is adjustable depending on processing conditions[11,12] and could be made comparable to the hole mobility of tetracene. For example, solution-derived ZTO films were reported to exhibit electron mobilities of 2.5 cm^2/Vs when annealed at 400 °C and 14 cm^2/Vs when annealed at 500 °C (Ref. 12) (although, from personal experience, we only managed a mobility range of 0.07 to 2.5 cm^2/Vs for the same annealing temperature range). Second, its low intrinsic carrier concentration ensures that the ZTO behaves as a semiconductor as opposed to a low work-function electrode. In the latter case, the device would act as a unipolar OLEFET, rather than an LH-LEFET. Third, it is easy to process from solution and mechanically robust—an added benefit that we detail in a subsequent paragraph.

The bottom-gate LH-LEFET was fabricated on clean p^+-Si/SiO_2 (200 nm thick dielectric) substrates. The ZTO precursor solution was prepared by dissolving $Zn(COOCH_3)_2$ and $SnCl_2$ (molar ratio 1:1) in 2-methoxyethanol at 0.3 M concentration. An equivalent molar ratio of acetylacetone was added to stabilize the mixture.[12] The solution was filtered and spin cast onto the substrate at 5000 rpm under nitrogen and annealed at 450 °C in air, which converted the acetate and chloride precursors to the amorphous oxide. The annealing temperature was chosen to yield a charge carrier mobility that better complements that of tetracene.

One half of the 20 nm-thick ZTO film was then protected with AZ5214 photoresist and the other half etched away in concentrated HCl to produce a clean edge where coverage ended. Once the photoresist was dissolved away and the ZTO film was washed and dried, tetracene (Sigma-Aldrich, sublimed grade) was sublimed onto the substrate (80 nm thick) through a pre-aligned shadow mask, which

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allowed a slight overlap of tetracene over the ZTO edge to ensure a complete interface between the two semiconductors. Subsequently, a new shadow mask was then aligned for sequential evaporation of Au/Ag source and drain contacts, with the tetracene/ZTO interface located in between the two contacts (the Ag layer on top acts only to thicken the Au contacts). The mechanical durability of ZTO is beneficial here because manual alignment of the shadow masks can cause scratches on softer semiconductor films. In our previous attempts using organic n-type materials, this scratching often led to unusable devices. The final device had a channel length of 200 μm and a width of 4 mm, and a schematic is provided in Figure 1, which also includes a simplified and approximate energy level diagram for reference.[13–15]

Electrical characterisation was performed under nitrogen using a Keithley 4200 connected to a micro-manipulator probe station. Unipolar tetracene and ZTO FETs fabricated under the same process conditions and on the same substrate as the tetracene/ZTO LH-LEFET exhibited a saturation hole mobility of ~0.071 cm^2/Vs and a saturation electron mobility of ~0.81 cm^2/Vs, respectively.

Transfer (drain current vs. gate voltage) curves of the tetracene/ZTO LH-LEFET are shown in Figure 2(a), at drain voltages of 0 to −100 V in 5 V steps. The transfer curves show a bell-shaped profile at gate voltages between −60 to −100 V, which is indicative of lateral heterostructure field-effect bipolar transistors.[9,16] The interested reader will find, in Ref. 17, a detailed analysis of this bell shape, but a summary of its origin is as follows. Under a negative drain voltage, high conductivity is induced in the n-type ZTO via the field effect when the gate voltage is low. In the p-type tetracene, however, the analogous hole-conducting channel is suppressed and the overall current remains low. As the gate voltage becomes more negative, formation of the hole-conducting channel in the tetracene becomes increasingly favourable and the current increases to a maximum. When the gate voltage is increased further, the electron-conducting channel in the ZTO is suppressed and the overall conductivity of the device, along with its current, falls.

The bell-shaped transfer curves are partially obscured by a large background current that occurs at gate voltages between 0 to −60 V. Measurement of the gate current reveals the background current to be composed mostly of current flowing between the drain and gate contacts, as shown in Figure 2(b). This parasitic gate current decreases with

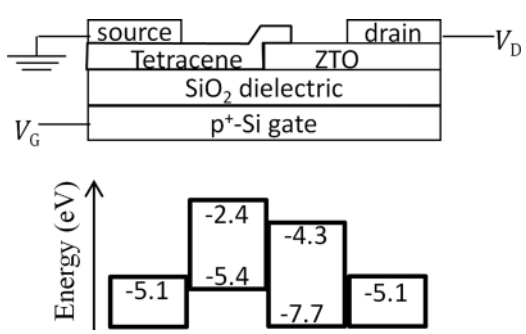

FIG. 1. Schematic diagram of the tetracene/ZTO LH-LEFET, showing the heterojunction within the transistor channel. A simplified diagram showing approximate energy levels of tetracene, ZTO, and Au.

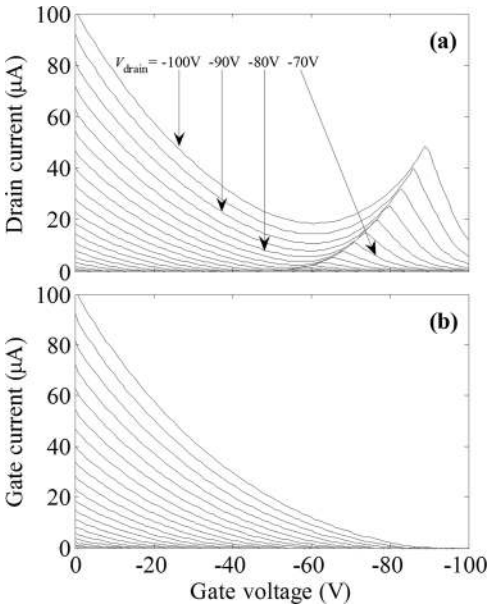

FIG. 2. (a) Transfer characteristics (drain current vs. gate voltage) of the tetracene/ZTO LH-LEFET at drain voltages of 0 to −100 V at 5 V steps. Between gate voltages of −60 and −100 V, the transfer curves are bell-shaped, a telling characteristic of lateral heterostructure bipolar FETs. (b) Gate current vs. gate voltage curves.

increasing gate voltage, suggesting a possible spreading current pathway through the ZTO due to a lateral field extending beyond the source-drain channel. Such a pathway is likely due to the combined effect of the contiguous, unpatterned ZTO film occupying half the substrate area and the presence of pinholes in an imperfect/damaged dielectric layer. On the other hand, the gate current is practically zero at very large negative gate voltages. In this regime, the tetracene is highly conductive, and the near-zero current indicates the absence of similar parasitic current pathways through the tetracene. This suggests that the gate current is wholly a drain-gate current without a source-gate component and allows for correction of the drain current data.

In Figure 3, the gate current has been subtracted from the drain current to better reveal the actual bell-shaped drain current. In addition, based on the corrected data, the output curves (drain current vs. drain voltage) have been plotted in the inset for gate voltages from 0 to −100 V in 5 V steps.

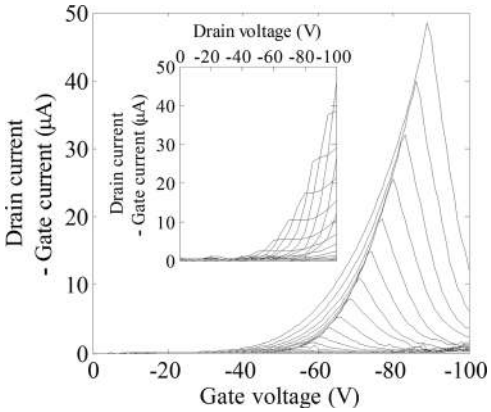

FIG. 3. Transfer curves of the LH-LEFET after correcting for the parasitic gate current, clearly showing the bell shape. (Inset) Corresponding output curves (drain current vs. drain voltage) from the corrected current data.

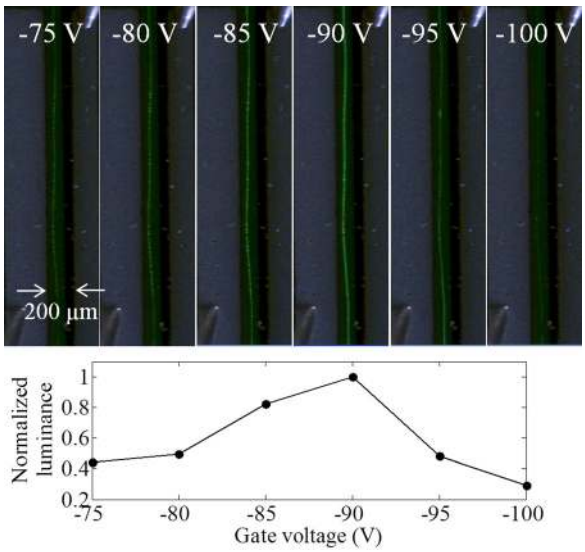

FIG. 4. (Color online) Optical photographs of the LH-LEFET under operation at a drain voltage of −100 V and gate voltages of −75 to −100 V. Going from left to right in each frame is the source contact (grey), tetracene (green), ZTO (black), and the drain contact (grey). The bright green emission appears to be in the middle of the tetracene side only because the tetracene overlaps the ZTO—a fabrication artifact. Emission is brightest at −90 V, which corresponds to the peak seen in the transfer characteristics in Figs. 2 and 3.

Both sets of curves closely resemble output and transfer curves in earlier reports of lateral heterostructure bipolar FETs.[9,16,17]

Figure 4 is a series of photographs showing the green light emission from the tetracene/ZTO LH-LEFET, while operating under a drain voltage of −100 V and gate voltages from −75 to −100 V. These photographs were captured using the CCD camera attached to the optical microscope of the probe station. Exposure time and other camera settings were kept constant. The green emission was also clearly visible to the naked eye in a darkened room.

Going from left to right within each frame, the different coloured bands represent the Au/Ag source contact (grey), tetracene (green), light emission (bright green), ZTO (black), and the Au/Ag drain contact (grey). At first glance, light appears to emit from within the tetracene side instead of at the tetracene/ZTO interface, which is the expected location; however, this is an only because the tetracene partially overlaps the ZTO, which is a fabrication artifact as described above.

From the CCD bitmap images, the brightness of a fixed section of the green emission was averaged to provide a normalised luminance (since camera exposure was kept the same for all six images), which is shown also in Figure 4. Qualitatively, light emission follows the trend of the transfer curve in Figure 3, increasing in brightness as gate voltage was increased from −75 V to a maximum at −90 V before decreasing as voltage was further pushed to −100 V. Unlike ambipolar OLEFETs,[18] the position of light emission in an LH-LEFET does not shift with gate voltage, since recombination takes place only at the heterojunction interface. It is highly likely that only a small fraction of emitted light escapes the device, due to trapping and absorption by the high-index Si substrate. Brightness could potentially be improved by using a substrate with lower refractive index or a reflective metallic gate substrate.

To conclude, we have demonstrated a working LH-LEFET based on tetracene and ZTO. The electrical characteristics of this device are typical of lateral heterostructure field effect transistors, and this is indicative of its bipolar nature. There are a few advantages in our approach to making an OLEFET: first, the lateral heterostructure architecture grants the device balanced carrier injection since, in principle, every carrier injected under steady state operation must recombine at the heterojunction. Second, amorphous oxide semiconductors such as ZTO have reliably good, adjustable electron mobilities—a relatively rare property in organic semiconductors. Also, low intrinsic carrier concentrations in ZTO ensure that the oxide semiconductor does not act as an electrode and nullify the first advantage. The architecture and the use of ZTO relaxes the requirements on the emissive organic semiconductor, which needs to have high hole mobility, but does not need to be ambipolar.

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