Light-Emitting Field-Effect Transistor Based on a Tetracene Thin Film

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We report the first organic light-emitting field-effect transistor. The device structure comprises interdigitated gold source and drain electrodes on a Si/SiO_2 substrate. A polycrystalline tetracene thin film is vacuum sublimated on the substrate forming the active layer of the device. Both holes and electrons are injected from the gold contacts into this layer leading to electroluminescence from the tetracene. The output characteristics, transfer characteristics, and the optical emission properties of the device are reported. A possible mechanism for electron injection is suggested.

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Introduction.—Since their discovery [1,2] organic field-effect transistors (OFETs) have been investigated extensively as demonstrated in recent review articles [3,4]. During the last ten years interest shifted from material related topics to morphology optimization, both for vacuum-deposited small molecules and polymeric semiconductors. The influence of substrate surface treatment [5] was also found to be of major importance to control the film morphology of the semiconductor, which determines the charge carrier mobility, trap distribution, etc. [6]. OFETs have been proven suitable to drive organic light-emitting diodes (OLEDs) [7,8], thus showing a promising device integration perspective.

Organic field-effect transistors are typically operated in unipolar accumulation mode, even though in principle ambipolar operation should be possible. The ambipolar operation mode may lead to carrier recombination, formation of excitons, and light emission. Such a light-emitting field-effect transistor would be the smallest possible integration of light source and switch, and has the additional advantage to allow control of the location of the charge recombination zone by means of applied electric fields. Provided it can be made from thin organic films, the organic light-emitting film transistor (O-LEFT) would constitute a crucial building block for applications in the optical information technology and nanotechnology. The idea of such a light-emitting transistor based on α-6*T* single crystals was reported recently [9]; however, the publication was subject to a scientific misconduct investigation [10] and finally withdrawn [11]. Since then, despite numerous attempts worldwide, light emission from an OFET, either based on single crystals or on thin films, has not been published.

Here we report the observation of light emission from an organic field-effect transistor based on a vacuum-deposited tetracene thin film. The organic light-emitting film transistor exhibits electrical characteristics similar to a unipolar OFET and its light emission intensity can be modulated by the gate and drain voltage. An explanation is proposed for the unexpected electron injection into tetracene from gold electrodes.

Experimental.—The O-LEFT devices were fabricated using heavily doped *n*-type silicon substrates (3–5 Ω cm resistivity) covered by a high quality thermally grown oxide with a thickness of about 285 nm acting as a gate electrode and a gate oxide, respectively. A thin chromium adhesive layer was deposited on the entire oxide surface before a 50 nm Au layer was deposited. The Au source and drain electrodes were photolithographically structured. They are configured as interdigitated fingers with a channel length of 5 μm and a channel width of 20 cm. The corresponding layout of the electrodes is displayed in Fig. 1. These substrate structures were then treated by the silane coupling agent *n*-octadecyldimethylchlorosilane to improve tetracene film homogeneity and substrate coverage. The treated substrates were directly transferred into an evaporation chamber by avoiding any exposure to ambient air. Tetracene (Chemos GmbH) was thermally evaporated at a base pressure of 1×10^{-6} mbar with a deposition rate of 5 Å/s onto the prestructured substrates.

The output and transfer characteristics of the O-LEFT were measured with an HP parameter analyzer (HP 4155A) and the light emission intensity was detected by a Si-photodiode placed about 2 mm above the emitting O-LEFT surface. The output photocurrent of the Si-photodiode was also fed into the parameter analyzer.

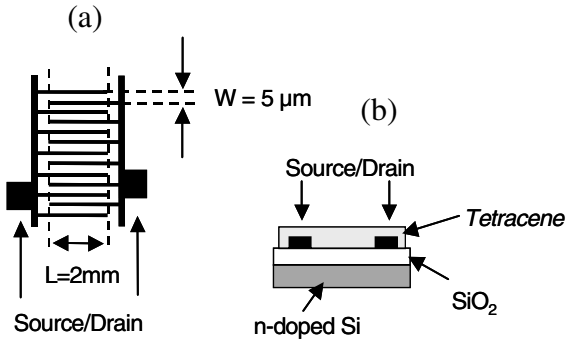

FIG. 1. Electrode configuration of the utilized O-LEFT: (a) interdigitated electrode structure of an O-LEFT, (b) cross section of two neighboring Au electrodes.

The electroluminescence spectrum of the O-LEFT was recorded with an optical simultaneous multichannel analyzer. A digital camera in combination with an optical microscope inside a glovebox was used to visualize the light emission of the operating transistor in order to determine the lateral position of the emission zone.

Results and discussion.—Typical output characteristics of an O-LEFT are displayed in Fig. 2. It exhibits the characteristics of a unipolar field-effect transistor with good saturation behavior. The transistor operates in the hole accumulation mode. The electrical transfer characteristics are displayed in Fig. 3. Its unusual saturation behavior is assigned to contact resistors on the source electrode as suggested by theoretical calculations [12]. Field-effect hole mobilities and threshold voltages, as derived from a fit to the experimental data in the linear regime of the $\sqrt{I_D}(V_{GS})$ plot of the transfer characteristics [3], are $\mu_h = 5 \times 10^{-2}$ cm^2/(V s) and $V_{th} = -25$ V, respectively, for $V_{GS} = -80$ V. On/off ratios in the range of 1×10^6 were also determined from the transfer characteristics. The threshold voltage of -25 V indicates a high concentration of trap states and suggests the presence of injection barriers. Indeed, the *S*-shaped output characteristics at small voltages indicate that even holes experience an injection barrier in reaching the tetracene highest occupied molecular orbital (HOMO) level from the Au electrode [12].

The electrical characteristics show no indication for ambipolar transport, neither was it possible to operate the transistor in pure *n*-type inversion mode. Obviously, the electron mobility is low and in addition the electron injection is weak. However, the significant light output at higher voltages confirms the injection of both electrons and holes. The optical characteristics of the same O-LEFT device as in Figs. 2 and 3 are displayed in Figs. 4 and 5. At first glance the optical response of the device is in agreement with expectations. The light emission associated with the electrical output characteristics is increasing with increasing source-drain voltages V_{DS} even at saturated drain currents (I_D). This is attributed to an augmented electron injection, which in turn leads to a better-balanced hole-to-electron ratio and hence to stronger luminescence. However, as shown by the saturation of the drain currents, any electron current has to be vanishingly small compared to the hole current. There is, however, an experimental fact that contradicts classical transistor theory, namely, the almost fixed onset of the light emission at $V_{DS} \approx -20$ V independent of the utilized gate voltage. Following thin-film transistor theory one would expect the onset of ambipolar operation for drain voltages exceeding the gate voltage [12]. In our devices, however (see Fig. 4) the onset of light output, resembling the onset of electron injection, starts at a common drain voltage of -20 V, even for gate voltages up to -80 V. In the present device only the intensity of the emitted light changes. A possible model for this atypical transistor behavior will be given at the end of the paper after having collected all experimental evidence. In order to prove that the observed light emission is not due to possible artifacts, such as dielectric breakdown phenomena and leakage currents through the gate oxide, the electroluminescence spectrum of the O-LEFT device has been recorded and compared to published charge recombination and exciton emission spectra in tetracene. The O-LEFT emission spectrum is shown in Fig. 6 and exhibits the typical spectral shape of tetracene thin-film emission centered at 540 nm [13–15]. The luminance was determined to be 45 Cd/m^2. However, this value can be considered only a rough estimate due to poor optical coupling of the light-emitting area to the luminance detection. The corresponding luminance efficiency of 0.0024 Cd/A shows that mainly holes account for the measured source-drain currents in the device containing only a small fraction of both charge carrier types that recombine radiatively. This also reveals the fact that electron injection is weak compared to hole injection.

The second possibility to generate light would be electrons leaking through the gate oxide and recombining with holes in the tetracene layer. To check this possibility the gate current was monitored parallel to all electric measurements. It was found in the μA range at zero drain voltage decreasing to half its value with increasing

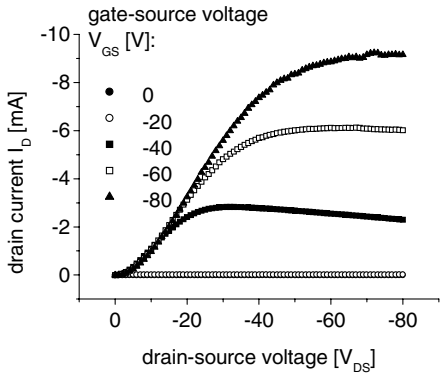

FIG. 2. Output characteristics of the tetracene O-LEFT for different gate-source voltages.

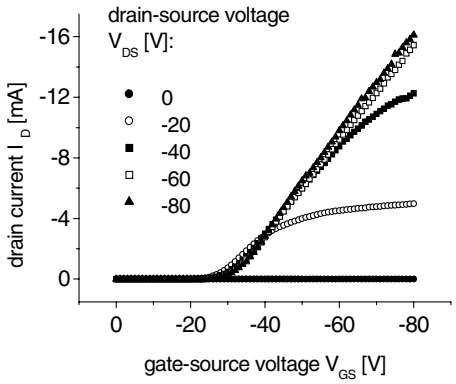

FIG. 3. Transfer characteristics of the tetracene O-LEFT for different drain-source voltages.

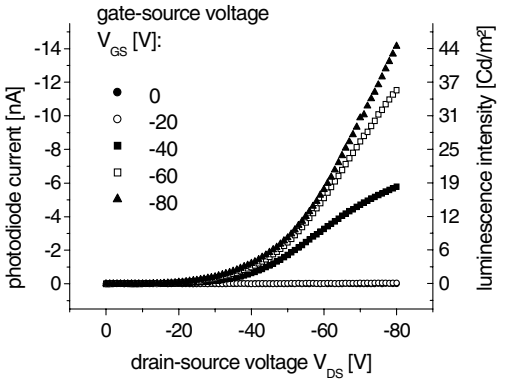

FIG. 4. Optical output characteristics of the tetracene O-LEFT as recorded by a photodiode for different gate-source voltages.

source-drain voltages. No correlation was observed with the onset of light emission. This fact alone makes leakage less likely as the origin for light emission, however, does not exclude the possibility. In case the observed leakage currents are hole currents, any additional electron currents could be small with respect to the total observed currents and therefore not observable.

To obtain an even deeper insight into the mechanism of light emission an optical image of the operating transistor was taken and is displayed in Fig. 7. One observes that the light is emitted only from locations close to the drain electrode. In addition the position of light emission is found to be independent of the applied voltage. This excludes beyond any doubt gate oxide leakage as the origin for light emission. In case of leakage the emission zone should be spread over the whole transistor channel. In addition, the highest light intensity should be found near the source electrode, where the electric field through the gate insulator is maximal.

Concerning the working mechanisms of the O-LEFT device some questions remain to be answered. A key aspect for understanding the device working principle

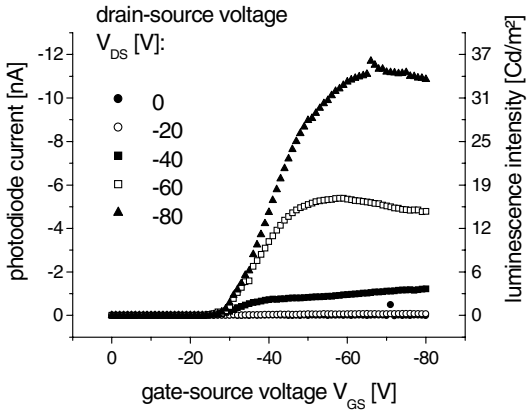

FIG. 5. Optical transfer characteristics of the tetracene O-LEFT as recorded by a photodiode for different drain-source voltages.

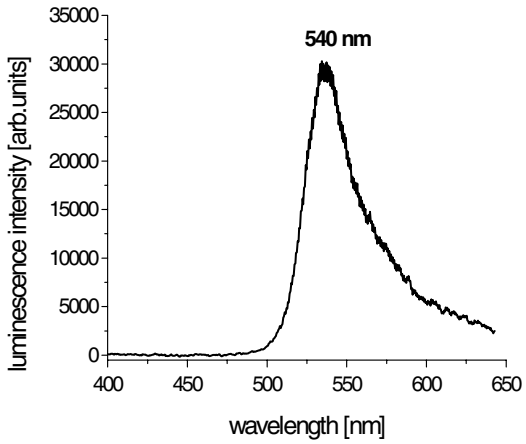

FIG. 6. Emission spectrum of the O-LEFT with $V_{GS} = -80$ V and $V_{DS} = -80$ V.

concerns the detailed mechanism of electron injection. As mentioned above, the transistor operates in a hole accumulation mode, which is a consequence of the relatively small injection barrier for holes from Au electrodes into tetracene. Reported values are 5.1 eV for the Au work function and 5.4 eV for the HOMO level of tetracene [16]. This results in a barrier height of 0.3 eV, which can easily be overcome at room temperature. Considering on the other side the energetic position of the lowest unoccupied molecular orbital (LUMO) level of tetracene at 2.4 eV [16], electrons have to surmount a barrier of 2.7 eV, which seems to be impossible by means of thermal energy alone. However, high electric fields may allow a tunneling through this barrier by field injection. The question at this point is still, where do these electrical fields come from, since classical transistor theory forbids electron injection.

There is one possible deviation from the ideal transistor device structure, which has not been taken into account so far. Such a modification may result from different injection mechanisms following from physical anomalies at the source or drain electrodes. Therefore the utilized electrodes were investigated by scanning electron microscopy and were found to be underetched up to 2 μm

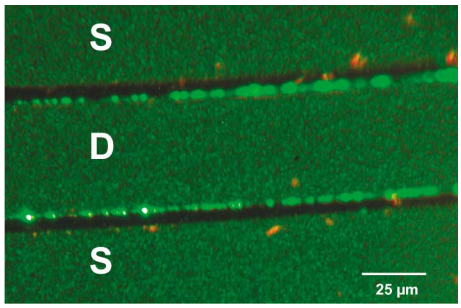

FIG. 7 (color). Picture of an operating O-LEFT taken through an optical microscope. Source (S) and drain (D) electrodes are arranged as shown in Fig. 1(a).

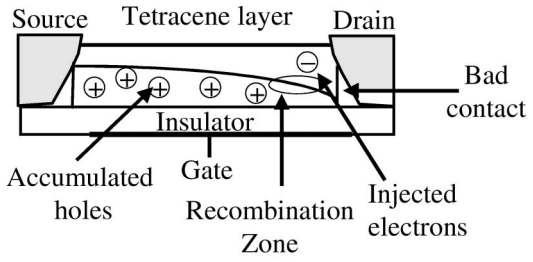

FIG. 8. O-LEFT in hole accumulation mode. Because of a bad contact at the drain electrode, electrons can be injected into tetracene far away from the gate dielectric interface.

exhibiting rough edges and airgaps between the gold electrode and the oxide surface. This underetching is a well-known problem for the electrochemical etching process during lithographic patterning. It is due to different etching processes utilized for the adhesive chromium layer and the gold layer on top. Such an electrode configuration is schematically presented in Fig. 8. The resulting shadowing effect during deposition of the tetracene thin film may cause bad electrical contacts between both source and drain electrodes and the dielectric interface of the gate oxide.

Taking these bad contacts into account the deviation of the characteristics of the present transistor from a common ambipolar transistor can be explained as follows: During operation the channel is filled with holes at the organic/dielectric interface (hole accumulation mode). However, due to the imperfections at the drain electrode, holes located near the gate insulator cannot directly reach the drain electrode due to an enlarged resistance of the bulk tetracene compared to the transistor channel. They have to traverse a certain distance through the tetracene bulk film before being able to reach the drain electrode. Since in this case the field of the gate electrode is mainly screened by the positive charge of the holes at the dielectric interface, a strong electric field is generated at the drain electrode, which may be high enough, especially on some kinks of the drain electrode, to allow injection of electrons by field emission [17]. This would explain why (1) the light emission on the drain electrode in Fig. 7 is very inhomogeneous, (2) no shift of the emission zone is observed with varying gate voltages, and (3) why electron injection is possible despite the transistor not operating in an ambipolar mode. Under such conditions the here-described light-emitting transistor is, in principle, a common OFET combined with an OLED where the transistor drain electrode takes the function of the OLED cathode and the OLED anode is missing, being replaced by the transistor channel.

Despite the fact that the here described O-LEFT devices are still in their embryonic state and not all mechanisms are understood in detail, the practical implications of this novel device can be potentially important. The O-LEFT, if properly designed, could replace the presently used combination of thin-film transistors with imaging devices such as liquid crystal displays or OLEDs. The advantage of the present design compared to any single crystal approach is the ability to vapor deposit the organic semiconductor, which can be performed on any kind of substrate, including plastic surfaces. There is also a good chance that O-LEFTs can be produced with other light-emitting materials including polymers.

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[1] F. Ebisawa, T. Kurokawa, and S. Nara, J. Appl. Phys. **54**, 3255 (1983).
[2] A. Tsumura, H. Koezuka, and T. Ando, Appl. Phys. Lett. **49**, 1210 (1986).
[3] G. Horowitz, Adv. Mater. **10**, 365 (1998).
[4] C. D. Dimitrakopoulos and P. R. L. Malenfant, Adv. Mater. **14**, 99 (2002).
[5] D. J. Gundlach, J. A. Nichols, L. Zhou, and T. N. Jackson, Appl. Phys. Lett. **80**, 2925 (2002).
[6] A. Hepp, N. von Malm, R. Schmechel, and H. von Seggern, Synth. Met. **138**, 201 (2003).
[7] A. Dodabalapur, Z. Bao, A. Makhija, J. G. Laquindanum, V. R. Raju, Y. Feng, H. E. Katz, and J. Rogers, Appl. Phys. Lett. **73**, 142 (1998).
[8] H. Sirringhaus, N. Tessler, and R. H. Friend, Science **280**, 1741 (1998).
[9] J. H. Schön, A. Dodabalapur, Ch. Kloc, and B. Batlogg, Science **290**, 963 (2000).
[10] Report of Investigation committee, Bell Labs, 2002.
[11] Z. Bao, B. Battlog, S. Berg, A. Dodabalapur, R. C. Haddon, H. Hwang, Ch. Kloc, H. Meng, and J. H. Schön, Science **298**, 961 (2002).
[12] R. Schmechel and Heinz von Seggern, in *Light Emitting Field-Effect Transistor: Simple Model and Underlying Functional Mechanisms*, SPIE Proceedings Vol. 48 (SPIE–International Society for Optical Engineering, Bellingham, WA, 2003).
[13] A. Wappelt, A. Bergmann, A. Napikowotzki, H. J. Eichler, H. J. Jüpner, A. Kummrow, A. Lau, and S. Woggon, J. Appl. Phys. **78**, 5192 (1995).
[14] H. Müller and H. Bässler, Chem. Phys. Lett. **36**, 312 (1975).
[15] Literature values for tetracene emission were validated by our own photo- and electroluminescence measurements on thin films.
[16] M. Pope and Ch. Swenberg, *Electronic Processes in Organic Crystals and Polymers* (Oxford University Press, New York, 1999), 2nd ed.
[17] K. Murata, S. Cinà, and N. C. Greenham, Appl. Phys. Lett. **79**, 1193 (2001).