Doped organic transistors operating in the inversion and depletion regime

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The inversion field-effect transistor is the basic device of modern microelectronics and is nowadays used more than a billion times on every state-of-the-art computer chip. In the future, this rigid technology will be complemented by flexible electronics produced at extremely low cost. Organic field-effect transistors have the potential to be the basic device for flexible electronics, but still need much improvement. In particular, despite more than 20 years of research, organic inversion mode transistors have not been reported so far. Here we discuss the first realization of organic inversion transistors and the optimization of organic depletion transistors by our organic doping technology. We show that the transistor parameters—in particular, the threshold voltage and the ON/OFF ratio—can be controlled by the doping concentration and the thickness of the transistor channel. Injection of minority carriers into the doped transistor channel is achieved by doped contacts, which allows forming an inversion layer.

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Controlled doping of semiconductors leads to highly reproducible transistors with precisely tunable device characteristics\(^1\), which is witnessed by the success of inorganic metal-oxide-semiconductor field-effect transistors. However, most surprisingly, organic field-effect transistors (OFETs) almost exclusively use electrically undoped semiconductors and OFETs with doped channels are rarely reported. Doping has only been used to realize depletion type OFETs\(^2\)–\(^{10}\). In these devices, doping either results in a low ON/OFF ratio for higher dopant concentrations, or the effect of doping on the transistor behaviour, for example, on the threshold voltage, is limited when lower concentrations are chosen\(^3\),\(^5\). In contrast to depletion type transistors organic inversion transistors, that is, transistors in which, for example, a p-inversion layer is created in an n-doped channel layer, have not been realized so far. The main challenge of reaching the inversion regime is the generation of minority charge carriers at the interface to the gate insulator. Scheinert et al.\(^{11}\) have argued that the inversion regime in organic metal-insulator-semiconductor (MIS) capacitors cannot be reached due to the large time constant of minority charge carrier generation. Scheinert et al.\(^{11}\) and Lindner et al.\(^{12}\) also proposed theoretically that minority charge carriers could be injected from the source of the OFET into the channel, if injection is efficient. However, Brondijk et al.\(^{13}\) recently showed that injection of minority charge carriers in P3HT-based transistors is too inefficient in order to generate an inversion channel.

Here, we report both inversion and depletion OFETs. Depletion OFETs with high ON/OFF ratio are realized by keeping the doped layer as thin as possible, that is, a few nanometres only, which allows for a full depletion of the doped region by the applied gate voltage. Organic inversion transistors are facilitated by additionally doping of the source and drain contact, which leads to efficient injection of charge carriers and allows for the creation of an inversion layer. We show that doping leads to a precise control of the threshold voltage of OFETs. Controlling and stabilizing the threshold voltage in OFETs is essential in the design and production of larger integrated circuits. In particular, small and symmetric threshold voltages for p- and n-type transistors are needed for low-power complementary circuits.

**Results**

**Doping of pentacene by the p-dopant F\(_6\)TCNQ and the n-dopant W\(_2\)(hpp)\(_4\).** Owing to its high hole mobility, pentacene is chosen as organic matrix material. F\(_6\)TCNQ is used as p-dopant, which shows a similar doping strength as the commonly used dopant F\(_3\)TCNQ. F\(_6\)TCNQ, however, has a higher molecular weight, which allows a more precise control during deposition run, for example, on the same substrate and at the same time constant of device field effect carrier generation. F\(_6\)TCNQ and W\(_2\)(hpp)\(_4\) are used, which is also well known to significantly increase the conductivity of the fullerene C\(_{60}\) (ref. 15).

To prove that pentacene is p- as well as n-dopable and to distinguish between trap filling by doping and real doping, that is, generation of free charges, we study the shift of the Fermi level upon doping by ultraviolet photoelectron spectroscopy (UPS).

Figure 1 shows the UPS spectrum for 45 nm of pentacene, which is either intrinsic, doped by 0.5 wt% or 1 wt% of the p-dopant F\(_6\)TCNQ, or doped by 0.5 wt% or 1 wt% of the n-dopant W\(_2\)(hpp)\(_4\). To avoid any reactions at the substrate surface (silver) and to make the growth of pentacene more reproducible, a 4 nm interlayer of MeO-TPD is included between the silver substrate and pentacene.

Figure 1 shows that the highest occupied molecular orbital (HOMO) states shift strongly towards the Fermi level (at \(0\) eV) for p-doping and away from the Fermi level for n-doping. The work function of the films and the difference between the Fermi level and the HOMO of pentacene \(\eta\) for the different doping concentrations are summarized in Table 1. For the intrinsic material, the Fermi level is close to the midgap position (\(\eta = 1.31\) eV, ionization potential = 4.94 eV). Already at a dopant concentration of 0.5 wt% doping leads to a strong Fermi level shift towards the HOMO for p-doping (\(\eta = 0.46\) eV) and away from the HOMO for n-doping (\(\eta = 2.05\) eV), whereas the ionization potential of pentacene remains almost unchanged. Further increase of the dopant concentration leads only to a weak additional shift of the Fermi level (\(\eta = 0.43\) eV for 1 wt% of p-doping and \(\eta = 2.12\) eV for 1 wt% of n-doping). These observations are in qualitative agreement with doping of the organic light-emitting diode (OLED) hole transport material MeO-TPD by F\(_6\)TCNQ\(^{16}\), F\(_6\)TCNQ or C\(_{60}\)F\(_3\) (ref. 17). Most importantly, the strong shift of the Fermi level towards the transport states indicates that free charges are generated. If doping would only fill traps the Fermi level would be pinned at the trap level, an effect observed, for example, at low concentrations in MeO-TPD\(^{17}\).

**Depletion transistors.** To realize depletion transistors, source and drain contact as well as the channel are p-doped. To keep the ON/OFF ratio of the intrinsic transistor, only a thin layer (6 nm) at the interface between gate oxide and pentacene layer is doped. The general set-up for the transistors is sketched in Fig. 2. They consist of aluminium gate electrodes, 120 nm of Al\(_2\)O\(_3\) dielectric layer deposited by atomic layer deposition (ALD), a thin p-doped layer of pentacene (6 nm, intrinsic, 0.5, 1 and 2 wt% F\(_6\)TCNQ), and 40 nm of intrinsic pentacene. The intrinsic layer ensures that the pentacene layer is decreased in later experiments. The source and drain contacts are defined by shadow masks and consist of 50 nm of p-doped pentacene (F\(_6\)TCNQ, 4 wt%) to ensure efficient injection and 50 nm of aluminium. Figure 3a shows the transfer characteristics of these transistors with varying doping concentrations in the channel. All devices are processed in a single deposition run, for example, on the same substrate and at...
Table 1 | Doping of pentacene studied by UPS.

| Dopant       | Doping concentration (wt %) | Work function* (eV) | η(eV) | Ionization potential† (eV) |
|--------------|-----------------------------|---------------------|-------|---------------------------|
| F6TCNNQ (p)  | 0.5                         | 3.63                | 1.31  | 4.94                      |
| F6TCNNQ (p)  | 1.0                         | 4.52                | 0.46  | 4.98                      |
| W2(hpp)2 (n) | 0.5                         | 2.94                | 2.05  | 4.99                      |
| W2(hpp)2 (n) | 1.0                         | 2.85                | 2.12  | 4.97                      |

*Work function, distance between transport states and Fermi level η. †Ionization potential for intrinsic and doped pentacene thin films.

Figure 2 | Doped OFETs. Set-up of the transistors consisting of Al2O3 as gate oxide, a thin doped layer of pentacene as transistor channel, an intrinsic pentacene layer and doped source and drain contacts.

eN
[44x419]gate oxide, a thin doped layer of pentacene as transistor channel, an

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concentration (2 wt%), the performance and in particular the sub-

samples to sample variations. Only at the highest doping con-

increased ON/OFF ratio, which, however, is most probably due to

the threshold voltage decreases. Most importantly, all doped

samples show the same ON/OFF ratio, that is, the doped layer can

be fully depleted. The 0.5 and 1 wt% sample even show a slightly

increased ON/OFF ratio, which, however, is most probably due to

sample to sample variations. Only at the highest doping concen-

tration (2 wt%), the performance and in particular the sub-

threshold swing of the transistor degrades. The devices show

moderate hole mobilities in the order of 0.1 cm²(Vs)⁻¹, which

can be explained by the high-k dielectric, which is known to

reduce the mobility in OFETs18, and by the deposition at room

temperature.

These results show that the threshold voltage can be controlled

by doping. The shift can be caused by two mechanisms: First,
doping increases the density of free holes in the channel region.

To reach the same density of free charges in the undoped

transistors, an additional voltage of \( \frac{eN_A d_{dop}}{C_{ox}} \) has to be

applied to the gate (\( N_A \), doping concentration; \( d_{dop} \), thickness of
doped film; \( C_{ox} \), capacitance of gate oxide; \( e \), elementary charge).

Thus, one can define the threshold voltage of the doped transistor

as (\( V_{FB} \), flatband voltage of transistors)

\[
V_{th} = V_{FB} + \frac{eN_A d_{dop}}{C_{ox}}.
\]

Alternatively, the shift of the threshold voltage can also be

explained by a filling of traps as proposed by Olothof et al.10 They

showed that doping at ultralow concentrations fill traps in the
type semiconductor \( C_{ox} \) reducing the threshold voltage in these
devices while keeping the ON/OFF ratio of the transistor10.

Similarly, Naab et al.19 showed that filling of traps in TIPS-

pentacene by an n-dopant facilitates a high electron mobility in

TIPS-pentacene (6,13-Bis(triisopropylsilylethynyl)pentacene).

However, the UPS results of p- and n-doped pentacene films

discussed above show that doping is effective. The Fermi Level

shifts strongly towards the transport states, even at relatively low

concentrations of 0.5–2 wt%, proving that free charge carriers are

generated in the bulk of pentacene.

Nevertheless, the shift of the flat band voltage could also be

caused by a filling of traps at the interface between gate oxide and

pentacene. To figure out if trap filling at the organic/gate oxide

interface has a significant effect on the threshold voltage, we vary

the thickness of the doped layer from 2 to 10 nm. Intentionally,

we keep the doping concentration at a high level of 2 wt% to

enhance the effect, which however leads to a slight deterioration

of the performance of individual transistors, in particular to a

reduction in mobility of pentacene to \(~0.06\) cm²(Vs)⁻¹. This effect can be explained by a structural phase transition of the

pentacene film from crystalline to amorphous20.

Figure 3b shows the result of this experiment. As expected, the

undoped reference device (black line) exhibits typical p-type

transistor behaviour with a threshold voltage of approximately

\(-3.6\) V. Higher positive voltages have to be applied to the gate to

fully deplete the transistor for increasing thickness of the doped

channel layer. For a 10 nm doped layer, a full depletion cannot be

reached any more, but for thinner layers the same off-current level as well as the same ON/OFF ratio of almost five orders of

magnitude are reached. This conclusion is in accordance with

previous studies of pentacene-based OFETs, where it was shown that carrier transport for undoped pentacene happens in the

few first pentacene/oxide interfacial layers21.

More importantly, the threshold voltage shifts with increasing

thickness of the doped layer. This shift is proportional to the

number of free charges in the doped layer (cf. equation (1)), which

is shown in Fig. 3c plotting the threshold voltage as
determined by a linear fit of Fig. 3b in the range of \(-10\) to \(-5\) V

versus the thickness of the doped layer. The threshold voltage shifts linearly from \(-3.6\) V for the undoped transistor to \(0.8\) V

for the 10-nm thick doped layer. Assuming a dielectric constant of

the Al2O3 layer of 7.8, a density of free charge carriers in the
doped layer of \ (~2.6 \times 10^{18} \) cm⁻³ is determined by equation (1).

This carrier density corresponds to a doping efficiency (density of

free holes divided by density of dopants) of 5.9%, which is in the

same order of magnitude as the doping efficiency observed in

other material systems16.

In summary, the linear shift of the threshold voltage with the

thickness of the doped layer clearly shows that the shift of

threshold voltage is not due to filling of traps at the organic/gate

oxide interface, but due to a real doping of free charges into the

channel.

Inversion transistors. In contrast to organic depletion transis-
tors, organic inversion transistors have not yet been reported in
Figure 3 | Organic depletion transistors. (a) Transfer characteristics of depletion transistors with a 6 nm p-doped channel at varying concentrations (0, 0.5, 1 and 2 wt% F6TCNNQ, $V_{DS} = -20$ V). The transfer characteristic is consistently shifted to the right and the threshold voltage decreases. In b, the thickness of the doped channel at a constant doping concentration (2 wt%) is varied. Thicker layers lead to a stronger shift of the threshold voltages plotted in part c. The thickness dependence of the threshold voltage shift proves that the effect is not due to trap filling at the oxide/organic interface.

As seen in Fig. 4b, the n-doped transistors clearly show a p-type characteristic, that is, the transistor turns on at negative voltages. In accordance with the CV characterization, the threshold voltage increases for increasing n-doping concentration.

This shift of the CV characteristic and the shift of the threshold voltage can be explained by an inversion of the n-doped layer at the oxide/pentacene interface. An increased potential has to be applied to the sample to first deplete the n-doped layer and to subsequently form an inversion layer of holes at even higher voltages. By increasing the density of n-dopants into the channel, more voltage has to be applied to the gate to reach inversion and the threshold voltage increases.

Discussion

Our results show that organic depletion and inversion transistors can be realized by doping a very thin layer of the organic semiconductor pentacene at the interface to the gate oxide. Doping leads to highly reproducible and tunable device characteristic. In particular, the threshold voltage of the transistors can be actively controlled by the thickness of the doped layer or the doping concentration while keeping other performance parameter such as the ON/OFF ratio, the sub-threshold swing and the mobility of pentacene constant. Owing to the precisely tunable device characteristics, we anticipate that doping in organic transistors will, similar to doping in inorganic transistors, become a key technology easing the design of larger integrated organic circuits.

Methods

Device fabrication. All samples are prepared by thermal evaporation in a single-chamber UHV tool (Kurt J. Lesker Company) at a base pressure of $10^{-7}$ mbar. The deposition system is equipped with a wedging tool, which allows depositing several samples with systematic variations on the same substrate in a single deposition run ensuring best comparability of the samples. Four different processing runs are

The intrinsic sample shows a depletion region at negative voltages and accumulation at positive voltages. N-doping the channel but keeping the hole injecting contact leads to a shift of the characteristic to more positive voltages. The shift of the characteristic is increased for increasing doping concentration. Changing from p-injecting contacts to n-injecting contacts allows observing accumulation of electrons as well as negative voltages (blue line in Fig. 4a).

Transistors built using the same stack show the same trends (cf. Fig. 4b). All samples have p-injecting contacts. The inversion transistors have a 6 nm thick n-doped channel. P-doped depletion transistors processed in the same run are shown for comparison as well.

the literature. The lack of inversion has been explained by inefficient injection of minority charge carriers into the inversion channel. However, in our design we have the possibility to selectively dope the source drain region and to facilitate efficient injection, for example, of holes into a p-type inversion channel formed in an n-doped channel region.

The formation of the inversion channel is studied by capacitance–voltage (CV) measurements as shown in Fig. 4a. The samples consist of aluminium back electrodes, 120 nm of Al$_2$O$_3$, a thin n-doped layer of pentacene (6 nm, intrinsic, 0.5, 1 and 2 wt% W$_2$(hpp)$_4$), 40 nm of intrinsic pentacene, 200 nm of p-doped pentacene (F$_6$TCNNQ, 4 wt%) to ensure efficient hole injection or 200 nm of n-doped pentacene (W$_2$(hpp)$_4$, 2 wt%) to allow for electron injection, and 50 nm of aluminium. The voltage is applied to the injecting contact and the back contact is kept at ground potential. All samples have been processed in a single deposition run to ensure highest reproducibility. Furthermore, care has been taken to characterize the different samples at identical conditions, that is, at the same maximum voltage, to make sure the shift of the characteristic is not due to different trapping/detrapping of doped electrons at the oxide/organic interface.

The intrinsic sample shows a depletion region at negative voltages and accumulation at positive voltages. N-doping the channel but keeping the hole injecting contact leads to a shift of the characteristic to more positive voltages. The shift of the characteristic is increased for increasing doping concentration. Changing from p-injecting contacts to n-injecting contacts allows observing accumulation of electrons as well as negative voltages (blue line in Fig. 4a).

Transistors built using the same stack show the same trends (cf. Fig. 4b). All samples have p-injecting contacts. The inversion transistors have a 6 nm thick n-doped channel. P-doped depletion transistors processed in the same run are shown for comparison as well.
Capacitors consisting of 120 nm Al$_2$O$_3$, a 6 nm n-doped pentacene film, an intrinsic pentacene layer and a hole (p-doped) injecting or electron (n-doped) injecting contact. Part a shows the transfer characteristic of transistors using the same gate stack and hole injecting contacts. For comparison, p-doped depletion transistors are shown as well.

Processed to reproduce the results, each consisting of 12 different transistors with slightly varied processing parameters. Despite some substrate to substrate variations, all runs show the same trends. The transistors show a small hysteresis between forward and backward sweep of 1–2 V only.

The samples consist of 60 nm Al as gate, 120 nm Al$_2$O$_3$ as gate oxide, a thin layer of pentacene (0–10 nm, Sensient, Wölln, Germany, density 1.32 g cm$^{-3}$, $M_{SW} = 278.35$ g mol$^{-1}$) doped either with the p-dopant F$_6$TCNNQ (2,2’-bis(5,6-diethyloxy)pyrimido[1,2-a]pyrimidinato)ditungsten (II), both Novaled AG. On top of the doped layer, 40 nm of intrinsic pentacene is deposited. The source and drain contact consist of a doped pentacene layer (50 nm (Fig. 3) or 200 nm (Fig. 4) doped with F$_6$TCNNQ at 4 wt%) and 50 nm Al. The bulky molecular dopants F$_6$TCNNQ and W$_2$(hpp)$_4$ are purchased from Novaled AG. As substrate a sputter-cleaned silver foil (99.995%, MatTeck) is used.

The UPS measurements are performed with a Phoibos 100 system (Specs) under ultra high vacuum conditions at a base pressure of 10$^{-10}$ mbar. Sample transfer without breaking the vacuum conditions is ensured by a direct connection of the UPS chamber to the evaporation tool. The energy resolution for UPS (HeI, 21.22 eV) is 130 meV and the experimental error (reproducibility) is estimated to be 50 meV. During the UPS measurement the sample is set to an acceleration potential of ~8 V. For each spectrum, the emission features due to secondary line excitations of the HeI discharge lamp are subtracted. For each sample the work function, defined as $W_f = 21.22$ eV – high binding energy cutoff (HBEC) as well as the HOMO – onset relatively to the Fermi level of the substrate are determined from the UPS spectrum. The measurements are kept as short as possible to avoid ultraviolet degradation of the organic materials.

UPS on doped layers. The UPS samples are prepared by thermal evaporation under high vacuum conditions (base pressure 10$^{-10}$ mbar) at room temperature in a multi-chamber tool. The doped layers are processed by co-evaporation of the matrix and dopant materials.

Characterization of the electrical characteristic of the transistors is done using the Semiconductor Parameter Analyzer Keithley 4200-SCS. All electrical measurements are performed at room temperature. All devices are stable under repeated cycling. To ensure a high comparability, all transfer characteristics of a series are measured at constant $V_{DS}$.

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Author contributions

B.L. designed the transistors, performed the measurements, wrote the manuscript and analysed most of the data. M.L.T. studied the doping effect in pentacene by UPS and analysed the UPS data. H.K. and A.Z. were involved in the sample preparation and made essential contributions to the interpretation of the data. C.H. and J.W.B. deposited, optimized and characterized the gate oxide. K.L. motivated and supervised this work.

Additional information

Competing financial interests: The authors declare no competing financial interests.

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