Pentacene organic field-effect phototransistor with memory-effect

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ABSTRACT

Now organic thin film transistor technology gains growing maturity, high performance organic photodetectors are the missing link towards full organic photosensitive sensor arrays, needed for the realization of applications like organic scanners and organic cameras. In the borderline of the research in organic solar cells, research in organic photodetectors has mainly been limited to organic photodiodes. However, phototransistors offer the possibility to reach higher sensitivities, thanks to the internal current gain of a transistor structure. This document focuses on organic field-effect phototransistors (organic photoFETs) where illumination can be used as an optical gate signal in addition to the electrical gate signal. An experimental description of pentacene photoFETs during broadband illumination is given. A distinction between the direct photocurrent and the additional current enhancement by a thresholdshift can be made, and a relation for this thresholdshift in function of time is revealed. Consequently, the difficulty to compare different organic photoFETs without any information about the illumination time is highlighted and a comparison based on this power law is proposed. Subsequently it is pointed out that by the stability of this thresholdshift after illumination, the pentacene photoFET does not only act as a light detector but also as a memory element, capable to store information about illumination power and duration for an extended amount of time.

Keywords: Image sensor, organic photoFET, thresholdshift, memory effect

1. INTRODUCTION

The extended interest in organic semiconductors is the result of several reasons.¹ Organic semiconductors can be processed on large area, flexible and transparent substrates because of their ease of manipulation at low temperatures. For the same reason organic devices have a potential cost advantage over their inorganic counterparts. In addition, the wide range in their molecular structure offers a substantial flexibility in the functionality of organic semiconductors. For example, the wavelength at which an organic emissive molecule emits light may be tuned by changing the side groups of the molecule. Currently, organic molecular and polymer semiconductors are used successfully to realize diodes,² field-effect transistors,³⁻⁵ basic circuits,^{6,7} light emitting devices,⁸ photovoltaic cells^{9,10} and photodetectors.¹¹⁻¹³

A highly sensitive photodetector is an essential element to obtain useful image sensors. Organic photodetectors are typically photodiodes, realized by sandwiching one or more appropriate organic layers between two conductive electrodes. The organic layers are optimized to maximize the absorption of photons, the dissociation of photogenerated excitons into free charge carriers and the collection of the free charge carriers at the contacts. Most challenging problem is the relatively fast annihilation of the excitons which restrict their dissociation efficiency into free charge carriers. Heterojunctions in a bulk or between thin layers are applied to by-pass this problem to a certain extent.^{9,10} Thin layers favour fast response times.¹² Accurate tuning of the layer thicknesses, taking into account their optical characteristics, may optimize the absorption of photons even

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in this thin layers.^{10, 13, 14} Nevertheless, there is no amplification of the photocurrent in these structures and optical-to-electrical gain is limited.

Less common is the organic bipolar phototransistor where the photocurrent is amplified by a current-to-current amplification. These devices have at least three organic layers, called emitter, base and collector, between two conductive electrodes. One type of photogenerated charge carriers is accumulated in the base layer by an appropriate energy level design. This accumulation facilitates the carrier injection from emitter to base and amplifies the current going through all three organic layers. Optimization of these devices includes the maximization of the enhanced injection from emitter to base. Gains greater than one are theoretically possible, but still limited in practice.¹⁵

In organic field-effect phototransistors (organic photoFETs), the transconductance of the field-effect transistor is used to amplify the photocurrent. Typical organic photoFETs have the same structure as organic field-effect transistors, with three conductive electrodes, source, drain and gate, and one organic semiconducting material. During illumination excitons are created in the bulk of the organic material and subsequently split into free charge carriers. One type of charge carriers is trapped in charge carrier traps within the structure of the device. In fact an organic photoFET is an integrating device building up a charge of trapped charge carries. This build-up gradually shifts the threshold voltage of the field-effect phototransistor, which is translated into an amplified drain current. To reach high gain an efficient photogeneration of free carriers, an excellent field-effect transistor characteristic and an efficient trapping of one type of charge carriers are needed. So far, the behaviour of an amplified drain current in an organic field-effect transistor under illumination has been noticed for various organic semiconductors.^{16–23}

Organic photoFETs, when operated in the off state of the field-effect transistor, show higher gains and signal-to-noise ratios compared to organic photodiodes and organic bipolar phototransistors. Moreover, thanks to their similar structure, the integration of organic field-effect transistors with organic photoFETs into a circuit is more straightforward than with other organic photodetectors. Consequently, organic photoFETs are promising device to realize a full-organic image sensors. Most works on organic photoFETs focuses on finding an expression for the photocurrent in function of the illumination power^{17–19, 24} but avoid to mention the illumination time. However, as an organic photoFET is an integrating device, different devices can not be compared without mentioning the illumination time. This works studies an organic photoFET based on pentacene, as this is an organic semiconductor providing excellent field-effect transistor characteristics^{25–27} and an high absorption in the visible spectrum (energy gap = 2.0 eV^{28, 29}), being two issues to obtain an efficient organic photoFET. An expression for the shift in thresholdvoltage in function of time is deduced. Consequently, the difficulty to interpret figures of merit of organic photoFETs is indicated.

2. FIGURES OF MERIT

Different figures of merit can be defined to quantify the characteristics of a photodetector. A first important figure of merit is the responsivity R expressed in A/W³⁰

$$R = \frac{I_{ph}}{P_{ill}} = \frac{I_l - I_d}{P_{ill}} \quad (1)$$

where I_{ph} is the photocurrent through the photodetector, P_{ill} the incident illumination power, I_l the current under illumination and I_d the current in the dark. The responsivity reveals to what extend the optical power is converted into an electrical current. The photocurrent can be written as a direct photocurrent $I_{ph,direct}$ multiplied by the optical-to-electrical gain G .³⁰

$$I_{ph} = I_{ph,direct} \cdot G = \eta_{IQE} \frac{qP_{ill}}{h\nu} \cdot G \quad (2)$$

Here, η_{IQE} is the internal quantum efficiency, representing the number of charge carriers photogenerated per photon. The direct photocurrent represents the photocurrent without any gain. The responsivity depends on the wavelength of the illumination source. To avoid this the external quantum efficiency η_{EQE} can be used.³⁰

$$\eta_{EQE} = \frac{\frac{I_{ph}}{q}}{\frac{P_{ill}}{h\nu}} \quad (3)$$

The relationship between these figures of merit can be clarified as follows

$$R = \eta_{IQE} \cdot G \cdot \frac{q}{h\nu} = \eta_{EQE} \cdot \frac{q}{h\nu} = \eta_{EQE} \cdot \frac{\lambda(nm)}{1240} \quad (4)$$

The internal quantum efficiency is smaller than or equal to one, but with a sufficient gain external quantum efficiencies greater than one can be obtained.

A second series of important figures of merit starts with the photoresponse $R_{l/d}$.¹⁷

$$R_{l/d} = \frac{I_l}{I_d} \quad (5)$$

As the noise current through the photodetector I_n depends on the dark current, the photoresponse can be related to the signal-to-noise ratio S/N which is defined by³¹

$$S/N = \frac{I_l}{I_n} \cong \frac{I_l}{I_d} = R_{l/d}. \quad (6)$$

A last series of figures of merit are the noise equivalent power NEP , the specific detectivity D^* and the dynamic range DR . The NEP is the incident power on the photodetector needed to generate a signal output equal to the noise output³²

$$NEP = \frac{I_n}{R} \quad (7)$$

and D^* is the reciprocal of the NEP , normalized to the detector area and the bandwidth³²

$$D^* = \frac{(A \cdot \Delta f)^{1/2}}{NEP}. \quad (8)$$

The dynamic range indicates the useful range for the power of the optical signal³³:

$$DR = 10 \cdot \log\left(\frac{P_{ill}^{sat}}{NEP}\right) \quad (9)$$

where P_{ill}^{sat} is the optical power saturating the photodetector.

In case of an organic photoFET, the photocurrent I_{ph} is the photocurrent through the drain electrode $I_{D,ph}$. As an organic photoFET is an integrating device, the photocurrent increases gradually under illumination. As a result, all these figures of merit are time dependent.

3. EXPERIMENT

3.1. Device Manufacturing Process

The structure of the device under study, is illustrated in Fig. 1 together with the chemical structure of pentacene. Interdigitated source and drain electrodes were fabricated in gold using a standard lift-off photolithography process, on the SiO₂ surface (100 nm en 30 nF/cm²) of a heavily doped silicon wafer, used as the gate electrode. Organic photoFETs with different sizes of transistor channel were realized and their dimensions are denoted as $W \times L$. Typical values for W and L are 1000 μ m and 10 μ m respectively. W is calculated as the channel width of one channel times the number of channels between the interdigitated source and drain electrodes. The number of channels was chosen so that the the overall shape of the interdigitated area was approximately a square. After the definition of the source and drain electrodes, the gold electrodes and the SiO₂ surface were respectively covered with an evaporated self-assembly monolayer of 1-dodecanethiol and octadecyltrichlorosilane (OTS), to improve the growth of the pentacene layer and to enhance the performance of the pentacene transistor.³⁴ Afterwards a pentacene thin film (~ 30 nm thick) was deposited under high vacuum ($\sim 10^{-8}$ Torr) at a deposition rate of 0.2-0.3 $\text{\AA}/\text{s}$ and a substrate temperature of 65 C. The pentacene, purchased from Aldrich, is purified twice by vacuum sublimation before loading into the deposition system.

3.2. Measurement Setup

The characteristics of the organic photoFETs were examined with a HP 4156C parameter analyser at a constant room temperature and in an nitrogen atmosphere. Currents down to 10^{-13} A could be measured. Broadband illumination in the visible spectrum was provided by a xenon lamp within the same nitrogen atmosphere. The devices were illuminated from the top (open) side, as shown in Fig. 1. The illumination power was measured using a Newport 818-UV Si photodetector with a calibration module.

Figure 1. (a) Cross-sectional view of a pentacene photoFET and (b) the chemical structure of pentacene.

4. RESULT AND DISCUSSION

To characterize the field-effect transistor behaviour of the fabricated pentacene photoFETs, the transfer characteristics were measured, and different device properties as mobility (μ_{FET}), current ratio of the on and off states (I_{on}/I_{off}), threshold voltage (V_T), inverse of the subthresholdslope (S^{-1}) and pinch-off voltage (V_{on}) were calculated. All transfer characteristics were measured with a double sweep from a negative to a positive gate-source voltage and back to a negative one. The device properties were calculated on the second part of the curve going from positive to negative gate-source voltages. All device showed excellent field-effect transistor characteristics, as illustrated in Fig. 2(a). The μ_{FET} and V_T of the photoFETs were obtained from the saturation regime as proposed by Horowitz,³

$$I_D^{sat} = \frac{W}{2L} \mu_{FET} C_i (V_{GS} - V_T)^2 \quad (10)$$

where L is the channel length, W the channel width, and C_i the capacitance per unit area of the gate dielectric layer ($C_i = 30$ nF/cm² for 100-nm-thick SiO₂). The S^{-1} was calculated as the slope of the I_D -vs- V_{GS} curve in saturation at V_{on} :

$$S^{-1} = \left. \frac{dV_{GS}}{d\log_{10}(|I_D|)} \right|_{V_{on}} \quad (11)$$

where V_{on} is arbitrarily defined as the gate-source voltage at which I_D reaches 1 nA. The calculated μ_{FET} , V_T and I_{on}/I_{off} are 0.60 cm²/(V.s), 1.2 V and $\sim 10^9$ respectively, as shown in Fig. 2(a). The S^{-1} and V_{on} are 0.24 V/decade and 4.3 V.

For the measurements of the characteristics of the device as a photodetector, the devices were measured under exposure to the white light of a xenon lamp. Fig. 2(b) shows the transfer characteristic of the pentacene photoFET before, during and after illumination. Two different effects of illumination can be distinguished.¹⁸ Before illumination, the current in the off state I_{off} is of the order of 10^{-13} A. During illumination, I_{off} is enhanced by an order of magnitude to $6 \cdot 10^{-12}$ A. After illumination, I_{off} drops back to its original value. Furthermore a clear shift in V_T and V_{on} towards more positive voltages can be noticed. As can be seen from the hysteresis in the double sweep curve under illumination, this shift is not stable during illumination. The second part of the curve, going from positive to negative gate-source voltage, has a more positive V_{on} than the first

Figure 2. (a) Transfer characteristic of the pentacene photoFET measured at $V_{DS} = -10$ V. (b) Transfer characteristic of the pentacene photFET before, during and after illumination. $V_{DS} = -10$ V and $P_{ill} = 0.2$ mW/cm².

part, which directs to a gradually shift during illumination. After illumination, V_T and V_{on} stay at an elevated level in comparison to the curves before illumination. The difference between the new and the original values is identical for V_T and V_{on} . Conversely the mobility and the subthresholdslope of the device before and after illumination are relatively unaffected, indicating that the electronic structure of the organic semiconductor is not affected by the illumination. During illumination, μ_{FET} and S^{-1} are slightly shifted which can be attributed to the deforming of the curves by the shift of V_T and V_{on} under illumination. These characteristics illustrate the additive effect of illumination to the gate-source voltage, both being able to control the density of charge carriers in the device. Either by a pulse of illumination or by applying a positive gate voltage, the field-effect transistor can be put in the on state.

In a pentacene photoFET under illumination, excitons are photogenerated in the bulk of the organic material and subsequently split into free charge carriers. The photogenerated holes give contribution to a direct photocurrent through the drain $I_{D,ph,direct}$ which can be particularly noticed in the cut-off region where $I_{D,ph,direct}$ is responsible for the increase of I_{off} . In saturation, $I_{D,ph,direct}$ is also present but small in comparison to the high on current. The shift in V_T and V_{on} can be attributed to the photogenerated electrons. These electrons are trapped in charge carrier traps, most likely at the interface between the pentacene and the OTS-treated SiO₂ surface where electron traps are known to exist.³⁵ These trapping of electrons screens the voltage present at the gate electrode, in this way effectively shifting the V_T and V_{on} of the device towards more positive voltages.³⁰ The effects of the photogenerated holes and the photogenerated electrons on the drain current are additive, so

$$I_D = I_{D,d} + I_{D,ph} = I_{D,d} + I_{D,ph,direct} + I_{D,ph,\Delta V_{on}} \quad (12)$$

where $I_{D,ph,direct}$ is the direct photocurrent from the holes and $I_{D,ph,\Delta V_{on}}$ is the photocurrent caused by the shift in V_T and V_{on} . From Fig. 2(b) it is clear that for a certain shift in V_{on} the highest photoresponse and responsivity are obtained for a gate-source voltage bias near to the initial pinch-off voltage $V_{on,init}$. The origin of this high sensitivities is not the direct photocurrent $I_{D,ph,direct}$ but the photocurrent $I_{D,ph,\Delta V_{on}}$ caused by the shift in V_T and V_{on} . As can be seen from fig. 2(b), a higher S^{-1} and I_{on}/I_{off} lead to a higher responsivity and photoresponse for an equal shift in V_{on} .

Obviously the direct photocurrent $I_{D,ph,direct}$ is direct proportional to the illumination power³⁰:

$$I_{D,ph,direct} = \eta_{IQE} \frac{qP_{ill}}{h\nu} \quad (13)$$

For the dependence of the shift in V_{on} and the corresponding photocurrent $I_{D,ph,\Delta V_{on}}$ on the illumination power, two different models can be found in literature. Noh et al.¹⁸ claim a logarithmic dependence

$$I_{D,ph,\Delta V_{on}} \sim \ln(1 + \eta_{IQE} \frac{qP_{ill}}{h\nu I_{D,d,n}}) \quad (14)$$

where $I_{D,d,n}$ is the dark current for electrons, while Hamilton et al.¹⁷ claim a power law relationship

$$I_{D,ph,\Delta V_{on}} \sim P_{ill}^\gamma \quad (15)$$

where γ is gate bias dependent and takes values between 0 and 0.8. Instead of measuring the dependence of $I_{D,ph,\Delta V_{on}}$ on illumination power, we measured the dependence on illumination time. Fig. 3(a) and fig. 3(b) show the drain current I_D for an illumination pulse of several tenths of seconds respectively for a gate-source bias $V_{GS} \approx V_{on,init}$ and a gate-source bias $V_{GS} > V_{on,init}$. The curves are measured on two different transistors on the same substrate. During illumination, the field-effect transistor was biased in the linear or cut-off region to obtain an uniform charge carrier density in the channel. For $V_{GS} \approx V_{on,init}$, $I_{D,ph}$ is a combination of $I_{D,ph,direct}$ and $I_{D,ph,\Delta V_{on}}$. For $V_{GS} > V_{on,init}$, in the first ten seconds there is only a contribution from $I_{D,ph,direct}$ in $I_{D,ph}$. Only when V_{on} is shifted till V_{GS} , there's also a contribution from $I_{D,ph,\Delta V_{on}}$ in $I_{D,ph}$. Measurements at more gate-source biases $> V_{on,init}$ not presented here, show that $I_{D,ph,direct}$ is independent from V_{GS} . Nevertheless, $I_{D,ph,direct}$ needs ~ 10 s to reach an equilibrium between the photogeneration of holes and the recombination of this holes with electrons. This explains the peak in $I_{D,ph,direct}$ in fig. 3(b) immediately after the start of the light pulse. From these curves the shift in V_{on} can be calculated. First, $I_{D,ph,\Delta V_{on}}$ is obtained by subtracting

Figure 3. I_D during an illumination pulse of several tenths of seconds for (a) $V_{GS} \approx V_{on,init}$ and (b) $V_{GS} > V_{on,init}$.

$I_{D,ph,direct}$, estimated as I_D during the first 10 s after the start of the illumination in fig. 3(b), from I_D . The current variation during illumination in the resulting curve, results only from the shift in V_{on} . By comparing each point in the resulting curve with the the original I_D -vs- V_{GS} curve in the dark, the V_{on} -shift at each point can be obtained. The result of this calculation for the two curves in fig. 3(a) and fig. 3(b) are presented in fig. 4(a). The first point in the curve for $V_{GS} > V_{on,init}$ is the point where V_{on} is shifted till V_{GS} . Both curves can be reasonably well fitted with a power law

$$\Delta V_{on} = C \cdot t^\gamma \quad (16)$$

where γ and C are gate bias dependent and $\gamma = 0.27$ and 0.41 and $C = 0.085$ and 0.065 for $V_{GS} \approx V_{on,init}$ and $V_{GS} > V_{on,init}$ respectively. The faster shift in V_{on} for $V_{GS} > V_{on,init}$ can be explained by the enhanced attraction of photogenerated electrons to the traps at the interface between the pentacene and the SiO_2 . As the

shift in V_{on} is both illumination time and gate bias dependent, the figures of merit of the pentacene photoFET are also dependent on these variables. In fig. 4(b) the drain current is shown during several illumination pulses of 50 s. As can be seen, after an illumination pulse, $I_{D,ph,direct}$ disappears within a few seconds, but I_D remains at an elevated level. In combination with fig. 2(b) we can conclude that the elevated I_D is caused by $I_{D,ph,\Delta V_{on}}$, and the trapped electrons are only relaxed with a time constant $\sim$ minutes.³⁶ The shift in V_{on} , and so the information about illumination power and illumination time, is stored for an extended amount of time within this device, which does not only act as a photodetector but also as a memory element.

Figure 4. (a) ΔV_{on} as a function of illumination time for $V_{GS} \approx V_{on,init}$ and $V_{GS} > V_{on,init}$. For both curves a fitting with a power law is shown. (b) I_D as a function of time during several light pulses of 50 s.

The responsivity and the photoresponse can now be calculated for the two cases of fig. 4(a). The responsivity and photoresponse due to the direct photocurrent is relatively independent of illumination time and gate-source bias V_{GS} . As the direct photocurrent is approximately 5×10^{-12} A, P_{ill} is 3.2 mW/cm^2 and the area of the transistor channel and the source and drain electrodes is $3 \times 10^{-4} \text{ cm}^2$, the responsivity can be calculated to be $5.2 \text{ } \mu\text{A/W}$ and the photoresponse to be 6. The responsivity and photoresponse due to the shift in V_{on} is gate-source bias and illumination time dependent. Assume a gate bias $V_{GS} \approx V_{on,init}$ V. After 1 s, V_{on} is shifted over 0.085 V. Since the inverse of the subthresholdslope S^{-1} is approximately 0.23 V/decade, this means a rise of 130% in I_D around $V_{on,init}$. As a result, the responsivity is only $1.4 \text{ } \mu\text{A/W}$, and the photoresponse is 2.3. So after 1 s, $I_{D,ph,direct}$ is dominating $I_{D,ph,\Delta V_{on}}$. Approximately 10 s of illumination time are needed for $I_{D,ph,\Delta V_{on}}$ to equal $I_{D,ph,direct}$. These values can be compared to the figures of merit for organic photoFETs found in literature as shown in table 1. At first sight our organic photoFETs seem to be under performing. However, in all these references, the figures of merit were calculated based on the transfer curves of the devices in dark and under illumination. If we do the same for our device on base of fig. 2(b), we obtain a responsivity of 7.5 A/W and a photoresponse of 3×10^8 . Bringing into account the use of a broadband illumination source in our experiments, our pentacene photoFETs belongs to the most efficient organic photoFETs reported. The enhanced figures of merit of our device with respect to the previous calculated values can be explained by a prolonged illumination time, one double sweep takes approximately 100 s, and the illumination at high positive gate biases, which seems to accelerate the shift of V_{on} . By this it is illustrated that interpreting figures of merit of organic photoFETs is difficult without information about the illumination time and the gate bias during illumination. In fact, the γ -factor of the power law relation between the shift in V_{on} and time as stated in this work, is a better variable to compare different devices, as this factor gives the direct relation between the shift in V_{on} , responsible for the most important share of the photocurrent, and the illumination time.

Table 1. Comparison of different organic photoFETs found in literature.

organic semiconductor	R [A/W]	$R_{l/d}$ [a.u.]	Illumination source
pentacene ¹⁹	50	1.3×10^5	365 nm, 5.0 mW/cm ²
pentacene ¹⁹	0.45	1000	650 nm, 5.0 mW/cm ²
pentacene ²⁰		10	540 nm, 0.1 mW/cm ²
copper phthalocyanine ¹⁹	2	3000	365 nm, 1.55 mW/cm ²
tetracene ²⁰		3000	450 nm, 0.1 mW/cm ²
F8T2* ²⁴	0.7×10^{-3}	100	white light, 270 mW/cm ²
F8T2 ¹⁷	2	1000	436 nm, 0.03 mW/cm ²
BAS-PPE ^{†21}	5.0×10^{-3}	6000	white light, 71 mW/cm ²
spiro compound ^{‡22}	1.3	500	370 nm, 0.13 mW/cm ²
BPTT ^{§18}	82	2×10^5	380 nm, 1.55 mW/cm ²

5. CONCLUSION

In conclusion, a photosensitive organic photoFET was fabricated based on pentacene, and the effect of broadband illumination on the device was studied. A distinction between a direct photocurrent supplied by photogenerated holes and a photocurrent originating in the shift of V_T and V_{on} by the trapping of photogenerated electrons could be made. The responsivity and photoresponse showed to be strongly time dependent and a power law relation between the shift in V_{on} and illumination time was revealed. Comparison with other organic photoFETs in literature showed to be difficult as mostly now information about illumination time can be found and a comparison based on the γ -factor of the revealed power law relation is proposed.

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*poly(6,9-dioctylfluorene-co-bithiophene)

†2,5-bis(dibutylaminostyryl)-1,4-phenylene-b-alkyne-b-1,4-bis(2-ethylhexyl)benzene

‡2,7-bis-(N,N'-diphenylamino)-2',7'-bis(bisphenyl-4-yl)-9,9'-spirobifluorene

§2,5-bis-bisphenyl-4-yl-thieno[3,2-b]thiophene

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