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Photoinduced negative magnetoresistance in 6,13-bis(triisopropylsilylethynyl)-pentacene field-effect transistors

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ABSTRACT

We report on photoinduced negative organic magnetoresistance in low external magnetic-fields (<100 mT) in 6,13-bis(triisopropylsilylethynyl)-pentacene (TIPS-Pentacene) field-effect transistors. An external magnetic field does not influence the dark current of our device. In contrast, there is a significant increase in photocurrent when magnetic field is applied to the irradiated device, which leads to negative magnetoresistance. The magnetoresistance and photoresponse values are strongly correlated and both are influenced by applied voltages and irradiation intensity. We attribute the observed photoinduced negative magnetoresistance in TIPS-Pentacene field-effect transistors to the presence of electron-hole pairs under irradiation. The overall dissociation probability of electron-hole pairs rises under the influence of an external magnetic field, which leads to a higher number of free charge carriers.

1. Introduction

Organic magnetoresistance (OMAR) is one main topic in modern organic electronics [1]. Due to its high values in low magnetic-fields measured at room temperature [2,3] OMAR is not only highly interesting for addressing fundamental questions about spin transport and spin manipulation in organic materials, but also for lightweight, flexible, and low-cost magneto optoelectronic applications. As an intrinsic effect OMAR exists in numerous organic semiconductors, from small molecules like Alq_3 [4–7], pentacene [4] and α-sexithiophene [8] to conjugated polymers like poly(3-hexylthiophene-2,5-diyl) [4], poly(9,9-dioctylfluorenyl-2,7-diyl) [4,9], poly(phenylene ethynelene) [4], and poly (*p*-phenylene vinylene)-derivatives [10–12]. OMAR is based on the magnetic-field dependent spin-correlation between quasiparticles that contribute to the current through the organic material and it is nearly always analyzed in two-terminal devices. For a deeper understanding of OMAR it is of high importance to explore this effect in organic field-effect transistors (FETs) because this (three-terminal) device geometry offers the possibility of pure unipolar charge transport. Until now there are only two examples of magnetic field-effects in organic FETs [13,14].

In this report we show photoinduced negative magnetoresistance (MR) in organic FETs based on 6,13-bis(triisopropylsilylethynyl)-pentacene (TIPS-Pentacene). In darkness the resistance is not influenced by magnetic fields. In contrast, a significant increase in photocurrent is observed when an external magnetic field is applied to the irradiated device. This magnetic-field induced photocurrent-enhancement leads to negative MR with values of up to 0.57% at 60 mT. The dependency of MR on the gate and drain voltages and its relationship to the magnetic-field and irradiation intensity will be presented. Finally, the possible mechanisms for negative MR in organic FETs will be described and discussed as well.

2. Experiment

The predefined bottom-contact organic FETs were purchased from Fraunhofer IPMS (Dresden, Germany) with channel lengths between 2.5 and 20 μm and channel width

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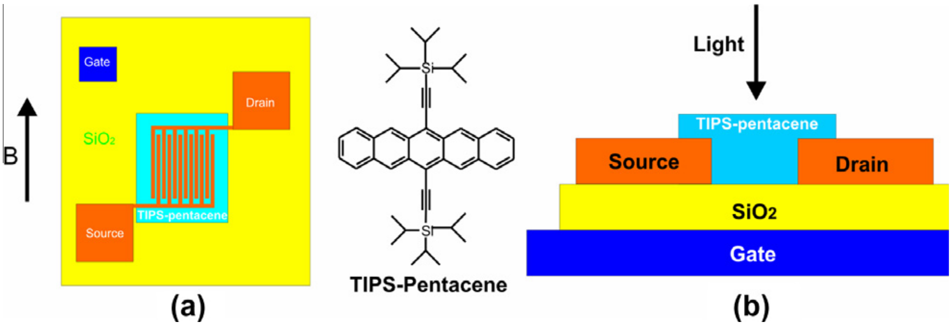

Fig. 1. (a) Top-view and (b) side-view of our experimental scheme. The magnetic field (B) can be either parallel or perpendicular to current flows between source and drain electrodes in the field-effect transistor. The illumination is directly done onto the surface of TIPS-Pentacene and is orthogonal to magnetic field and the current flow between source and drain electrodes. The chemical structure of TIPS-Pentacene is shown as well.

of 10 mm. The gate oxide SiO_2 is 230 ± 10 nm thick and the source and drain electrodes consist of 30 nm Au with 10 nm ITO as adhesion layer. TIPS-Pentacene ($\geqslant$99%) was purchased from Aldrich and used without further purification. Prior to the deposition of TIPS-Pentacene, the predefined substrates were cleaned with acetone, 2-propanol and deionized water, followed by oxygen-plasma treatment (300 W, 3 min) and exposure to hexamethyldisilazane to replace the natural hydroxyl end group of SiO_2 with an apolar methoxy group. Finally, TIPS-Pentacene was deposited by thermal evaporation at a base pressure of 3×10^{-7} Torr ($T_{\text{substrate}} = 298$ K, deposition rate = 0.66 Å/s) with a thickness of 78 nm. From the vacuum chamber the samples were directly transferred to a glove box (O_2, $H_2O < 0.1$ ppm) and placed in a homebuilt sample holder. This sample holder was mounted between the poles of an electromagnet with the magnetic field perpendicular or parallel to the direction of the current flow in organic FETs, as shown in Fig. 1. The magnetic field was varied between −86 mT and +86 mT. Current–voltage measurements were performed by using a Keithley 4200 semiconductor characterization system equipped with preamplifiers for low-current measurements. All measurements were performed at room temperature (≈298 K). The TIPS-Pentacene surface was irradiated at a wavelength of 671 ± 7 nm by using a Tungsten-Halogen Lamp (Spindler & Hoyer, Germany) in combination with an optical filter (Spindler & Hoyer, Germany). The diameter of the light spot on the surface of organic FETs was 4.5 cm and the variation in irradiation intensity was obtained by using KODAK neutral density filters. The baseline correction was done by using the peak-fitting module of the data analyzing software Origin 7.5.

3. Results and discussion

Fig. 2(a) contains the plots of drain current I_{DS} versus drain voltage V_{DS} at different gate voltages V_G (output characteristic) measured in darkness, under irradiation (296 μW) and under irradiation with applied external magnetic field (60 mT). In all our output characteristics we exclusively detected *p*-channel activity. Therefore, holes are the major charge carriers in the conducting channel. Fig. 2(b) is the zoom part of the output characteristics measured at $V_G = -20$ V and displays that a significant magnetic-field induced increase in photocurrent exists in TIPS-Pentacene FETs. This reduction in resistance leads to negative MR. Fig. 2(c) and (d) depicts transfer characteristics ($I_{DS} - V_G$) measured at $V_{DS} = -2$ and -30 V, respectively. The charge carrier mobilities were calculated in the saturation regime ($V_{DS} = -30$ V) by using $I_{DS} = (\mu WC/2L)(V_G – V_T)^2$, where L is the channel length, W is the channel width, C is the capacitance per unit area of the gate insulator, V_T is the threshold voltage, and μ is the charge carrier mobility. Under three different experimental conditions the charge carrier mobility and the fitted threshold voltage only change in a substantial way for the dark and the irradiated device. An external magnetic-field does not influence the charge carrier mobility at all and only has a very small impact on the threshold voltage. The charge carrier mobilities are 6.0×10^{-3} cm^2/Vs (in darkness), 6.2×10^{-3} cm^2/Vs (under irradiation) and 6.2×10^{-3} cm^2/Vs (under irradiation and applied magnetic field), respectively. A similar tendency is obtained for V_T: 2.50 V (in darkness), 2.53 V (under irradiation) and 2.54 V (under irradiation and applied magnetic field), respectively. However, the hole mobility in TIPS-Pentacene obtained in our experiment is approximately three times lower than that reported in literature [15]. Nevertheless, our TIPS-Pentacene FETs show an ON/OFF ratio of 10^6–10^7. Park et al. reported that the irradiation of TIPS-Pentacene FETs with 30 mW He–Ne laser ($\lambda = 632.8$ nm) produces a strong increase in the drain current by a factor of 10^4 and in mobility by a factor of 5 [15]. However, these results cannot be directly compared to our experiment because we used different conditions such as an irradiation wavelength of 671 nm with an intensity of up to 0.3 mW.

To quantify the influence of an external magnetic field on irradiated organic FETs, we determine the MR values of the samples under different voltage, irradiation intensity and magnetic-field strengths. However, we will not directly estimate the MR values from the output or transfer characteristics, but from time dependence of I_{DS}. Fig. 3 depicts MR in TIPS-Pentacene FETs recorded at $V_{DS} = -2$ V and $V_G = 0$ V. After 60 s, the light source was switched on and I_{DS} increased significantly, as displayed in Fig. 3(a). This photoresponse is defined as the ratio of the dark current and the current under irradiation. After that the external magnetic field was switched on and off three times in a series. Then, the light source was switched off

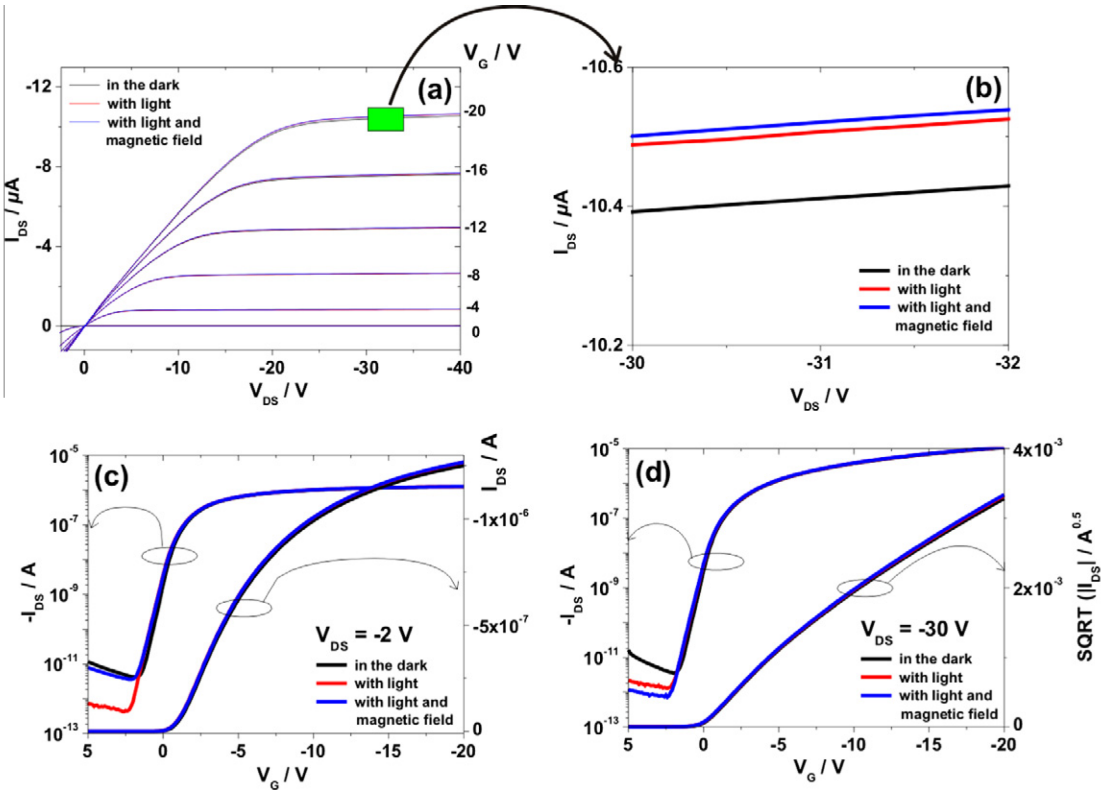

Fig. 2. (a) Output characteristics of a TIPS-Pentacene FET ($W = 10$ mm, $L = 20$ μm) and transfer characteristics in (c) linear ($V_{DS} = -2$ V) and (d) saturation ($V_{DS} = -30$ V) regimes, measured in the dark, under irradiation (296 μW) and under irradiation (296 μW) and magnetic field (60 mT), respectively. (b) The zoom part of output characteristics, measured at $V_G = -20$ V.

and the I_{DS}–time curve was obtained, as displayed in Fig. 3(a). This curve was used for the calculation of MR with MR = $\{[R(B) - R(0)]/R(0)\} \times 100\%$. $R(B)$ and $R(0)$ are resistances with and without applied magnetic-field, and are defined as $V_{DS}/I_{DS}{}^*$ and V_{DS}/I_{DS0}. With constant V_{DS} the MR is determined by $I_{DS}{}^*$ and I_{DS0}, where $I_{DS}{}^*$ is the photocurrent under influence of magnetic field and I_{DS0} is the photocurrent at zero magnetic field. Fig. 3(b) displays the zoom part 1 of Fig. 3(a) and contains the influence of a magnetic field on the devices in absence of irradiation. No significant change in current has been observed, indicating that a magnetic field does not change the resistance in darkness. In contrast, a significant magnetic-field effect is observed under irradiation, as shown in Fig. 3(c), which is the zoom part 2 of Fig. 3(a). After baseline-correction of bias stress caused drift, we obtained Fig. 3(d), which shows the magnetic-field induced photocurrent-change ($I_{DS}{}^* - I_{DS0}$) of the irradiated devices. The baseline-correction was done by linearly fitting the current without a magnetic field and subtracting this baseline function from the experimental data. We also observe that MRs do not depend on the direction of magnetic field with respect to the direction of current flow in organic FETs.

Fig. 4 displays the MR dependence on V_G and V_{DS}. V_{DS} (at constant V_G) does not influence the MR and photocurrent values, as shown in Fig. 4(a). In contrast, Fig. 4(b) shows that MR and the photoresponse strongly depend on V_G (at constant V_{DS}). In both cases the MR and photoresponse values are strongly correlated. If the photocurrent remains unchanged the MR values stay constant (Fig. 4(a)), but with decreasing photoeffect the influence of the magnetic field on the overall-current drops (Fig. 4(b)). Upon illumination two different processes take place in the conducting channel of the transistor, namely photoconductivity and photovoltaic effect [16,17], which leads to an increase in current of up to 25%. This value is comparable to that measured in pentacene phototransistor [14]. While photoconductivity occurs in the OFF-state at $V_G \approx V_{to}$, the photovoltaic effect dominates in the ON-state at $V_G > V_{to}$, with V_{to} being the turn-on voltage. Fig. 4(b) depicts, that a significant MR is observed at $V_G \approx V_{to}$ which rapidly decreases for $V_G > V_{to}$. Therefore, photoconductivity plays an important role, particularly if only a low density of gate-induced charges is present in depletion region ($|V_G| \approx |V_{to}|$). In this regime, the dark current has only a minor contribution to the overall current and the magnetic-field induced increase in photocurrent is observable. For $V_G > V_{to}$ the conduction is dominated by the gate-induced dark current, which is not sensitive to magnetic fields. Therefore, the magnetic-field induced modulations of the photocurrent becomes insignificant compared to gate-induced dark current and cannot be detected anymore.

Fig. 5 displays MR values and the photoresponse as a function of the irradiation intensity. For both measured voltages ($V_{DS} = -2$ V; $V_{DS} = -30$ V) exists a strong correlation between the MR values and the photoresponse for

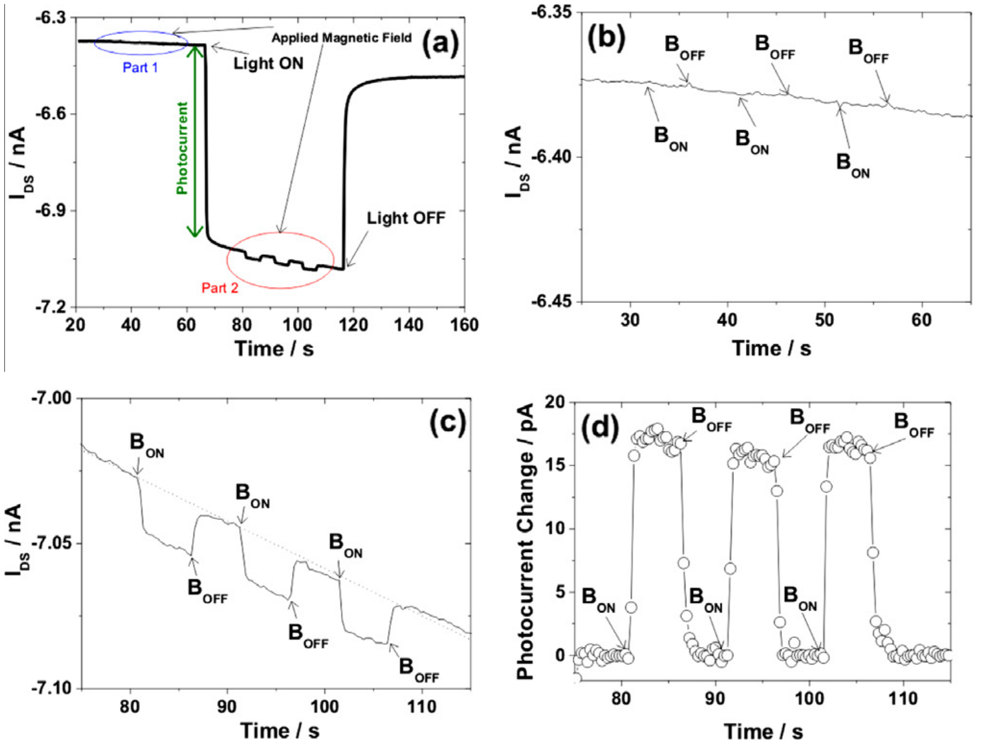

Fig. 3. (a) Time dependence of photocurrents in TIPS-Pentacene FET recorded at $V_{DS} = -2$ V and $V_G = 0$ V, measured under influence of irradiation (296 μW) and external magnetic field (60 mT). (b) Zoom-view of part 1 and it belongs to the dark current measured under influence of magnetic field. (c) Zoom-view of part 2 and it belongs to the photocurrent measured under influence of magnetic field. The dotted line was used as baseline for calculation of photocurrent change due to applied magnetic field. (d) The corresponding photocurrent change of part 2.

all measured irradiation intensities. For lesser irradiation intensities, the photoresponse and the associated MR values decrease in a nonlinear way. Therefore, the maximum MR value of −0.57% is observed at the maximum irradiation intensity of 296 μW (at a magnetic field of 60 mT). We also measured and calculated MR at different magnetic fields and the data was fitted by using empirical laws Non-Lorentzian, MR (%) = $\Delta R/R$ (%) $\propto B^2/(B + B_0)^2$, and Lorentzian line shapes, MR (%) = $\Delta R/R$ (%) $\propto B^2/(B^2 + B_0^2)$, as displayed in Fig. 6. The resulting fitting parameters are $B_0 = (22 \pm 3)$ mT for Non-Lorentzian and $B_0 = (32 \pm 13)$ mT for Lorentzian line shapes, respectively. Moreover, Non-Lorentzian line shape fitted our data a little bit better than Lorentzian line shape, which is similar as observed for irradiated FETs based on pentacene [14]. Our result is similar as reported for irradiated organic diodes ITO/*N,N′-di(naphthalene)-N,N′*-diphenyl-benzidine (NPB)/Al [18]. In this case, no magnetic field effect was observed when device was measured in darkness. In contrast, significant MR was measured as the device was irradiated. The magneto photocurrent at a bias smaller than the turn-on voltage can be well fitted by using Non-Lorentzian line shape, giving a high field component B_0 of ~140 mT. However, at applied bias larger than the turn-on voltage a new low field component B_0 of ~8 mT appears while the high field component remains. This low field component is well fitted by using Lorentzian line shape. Therefore, MR was observed in both irradiated organic diodes and organic FETs, but the field component B_0 is different due to different materials. In irradiated organic FET we do not observe high or low field component and the MR can be observed at gate voltage near or lower than the turn-on voltage. Furthermore, in organic diode the origin of Lorentzian line shape can be understood from the Hamiltonians for both hyperfine and spin–orbit interactions [19,20]. However, the Non-Lorentzian line shape behavior cannot be deduced from the hyperfine Hamiltonian. We obtained that both line shapes can fit our data, but the Non-Lorentzian line shape fitted the data slightly better than the Lorentzian one. However, the difference between both line shapes is too small to work out reliable physical conclusions.

All OMAR models are based on the magnetic field dependent spin-correlation between quasiparticles that contribute to the current of the device. In the following we will summarize several explanations for negative MR obtained in organic diodes.

(i) Due to their long lifetime triplet excitons have a high probability of transferring their excitation-energy to a charge carrier (polaron) by recombining to the ground state via so called exciton–charge interaction [21]. In exciton–charge-interaction model a reduction of the mobility of the polarons by scattering on triplet excitons is considered [22,23]. Under the assumption of a magnetic field dependent intersystem crossing rate, an external magnetic field can

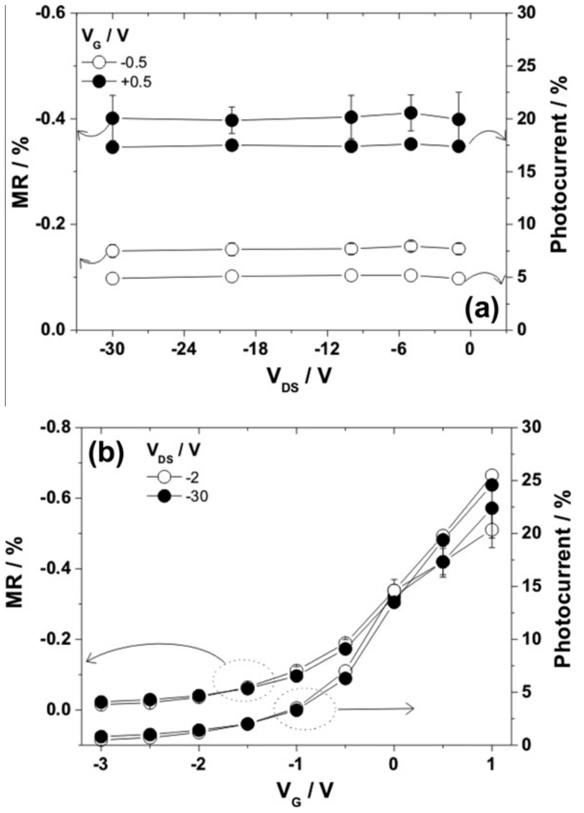

Fig. 4. (a) MRs plotted as a function of V_{DS} at $V_G = -0.5$ V and 0.5 V, respectively. (b) MRs plotted as a function of V_G at $V_{DS} = -2$ V and -30 V, respectively. The light intensity and the magnetic field were 296 μW and 60 mT, respectively.

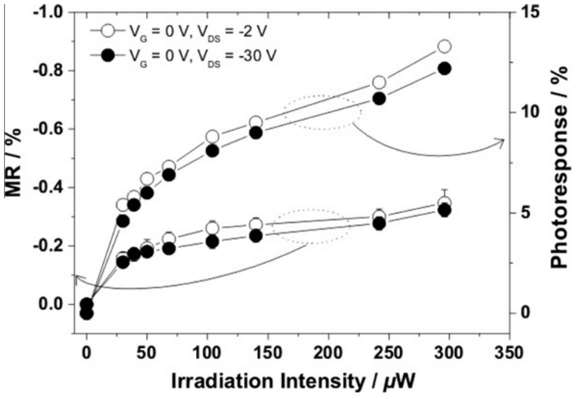

Fig. 5. MRs and photoresponses plotted as a function of irradiation intensity at $V_{DS} = -2$ V and -30 V. The V_G and the magnetic field were 0 V and 60 mT, respectively.

reduce the number of triplet excitons. This leads to less scattering events of free carriers and therefore an increase in mobility, which results in negative MR.

(ii) Negative MR has been observed in poly(9,9-dioctyl-1,4-fluorenylenevinylene) and poly(9,9-dioctyl-2,7-fluorenylenevinylene) [24]. The rise in current is due to the release of trapped charges by an external magnetic field. This also causes a change in the optical absorption in the low-energy range, which can be associated with deep traps [24].

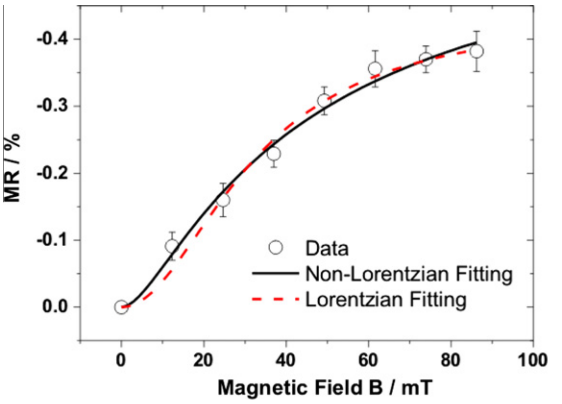

Fig. 6. MRs plotted as a function of magnetic field, measured at $V_{DS} = -2$ V and $V_G = 0$ V and incident light of 296 μW. The solid and dotted lines represent the fitting of the experimental data with non-Lorentzian and Lorentzian line shape, respectively.

(iii) The bipolaron model can explain MR in single-carrier devices [25,26]. It relies on the spin dependent formation of double occupancy of a particular site during hopping charge transport in organic semiconductors. This bipolaron formation is magnetic field dependent and the total number of bipolarons is reduced by an applied field [25]. If a considerably large amount of doubly occupied sites is available, there are less free carriers that can contribute to the current. By applying a magnetic field, the number of bipolarons is decreased and the number of free carriers is increased, which leads to negative MR [25].

(iv) Electron–hole (*e*–*h*) pairs consist of weakly coulomb-correlated oppositely charged, spatially separated polarons. Due to small exchange interaction the spin configuration of *e*–*h* pairs is flexible and their presence can lead to organic magnetoresistance [3,7,8,12,19,26,27]. One important pathway to *e*–*h* pairs is through relaxation from photoexcited states. Photon absorption in thin organic films results in excited quasiparticles, which consist of strongly coulomb-bound polarons with a short binding radius of less than 1 nm [28,29]. These so called (photoexcited) excitons may undergo a relaxation process to become *e*–*h* pair states with a longer electron–hole separation distance and less binding energy [28,29]. These can either recombine back into electrically neutral excitons (with rates r_S and r_T) or contribute to the current flow by dissociating to free polarons (with rates d_S and d_T). The *e*–*h* pairs can either be in a singlet or triplet spin configuration and the relative population of the two spin states can be modified by an external magnetic field [12]. Without a magnetic field the singlet level with the entire triplet manifold are degenerate and all four spin states are mixed via hyperfine interaction. In a magnetic field the Zeeman splitting lifts the degeneracy of the triplet levels and mixing between the singlet and triplet states is

> reduced. Prigodin et al. assumed that singlets have a larger recombination rate than triplets ($r_S > r_T$) and a shorter lifetime [7,8]. In the presence of a magnetic field the spin state mixing and the interconversion probability of triplets into singlets is reduced. This leads to more dissociation to free carriers, a higher current and negative MR [7,8]. Another process that can lead to negative MR was proposed by Hu et al. [12,28,29]. The authors point out that an external magnetic field increases the number of singlet *e–h* pairs through a field-dependent intersystem crossing rate [12,30,31]. Due to the stronger ionic character of singlet *e–h* pairs, they are stronger coupled to the ionic products of the dissociation-reaction and, therefore, singlet *e–h* pairs can be more easily separated into free charge carriers than triplet *e–h* pairs [12,32–34]. Hence, an external magnetic field boosts the number of singlet *e–h* pairs, which leads to the generation of additional secondary charge carriers by dissociation and it results in a negative MR [12].

The existing models have been developed to describe magnetic field-effects in vertical two-terminal devices such as organic light-emitting diodes or organic solar cells [4,29]. To be able to distinguish between the models based on unipolar or bipolar charge transport and to clarify the details of the underlying fundamental processes it is of high importance to realize true unipolar charge transport properties [35]. In vertical two-terminal devices this can be achieved by carefully minimizing the injection barrier for one charge carrier type and maximizing the injection barrier for the other [36]. Due to interfacial effects such as charge transfer or energy level alignment or formation of a dipole layer at organic/metal and metal/organic interfaces this is not an easy task [37]. In addition to the energy mismatch between the effective electrode work function and the energy of the transport level of the organic semiconductor we can further define the major charge carrier type by using gate voltage in three-terminal devices. Due to the small hole injection barrier between gold electrodes and the hole transport level of TIPS-Pentacene we find true hole transport in TIPS-Pentacene [38–40]. This is supported by our output characteristics that show only hole transport for all applied voltages (see Fig. 2).

Because TIPS-Pentacene FETs are completely built up of nonmagnetic materials, without any spin polarization, the negative MR can be attributed to intrinsic magnetic-field induced modulations of the charge carrier density n and/or the charge carrier mobility μ. In general MR can evolve from two different channels as described by $\mathrm{MR} = \mathrm{MR}(n) + \mathrm{MR}(\mu)$ [28]. As the excited-states-based channel, MR(n) is related to the magnetic-field-sensitive generation of secondary charge carriers from excited states [28]. Thereby the photoinduced excitation generates singlet excitons. The charge-transport-based channel MR(μ) depends on the existence of different types of charge carriers with unequal mobilities and a magnetic-field-dependent reaction path for intertype conversion [28]. Since the dark current in our true hole only device remains unaltered in an external magnetic-field and no magnetic-field-induced mobility-change in irradiated TIPS-Pentacene FETs could be observed, we can exclude the charge-transport-based channel MR(μ) as the underlying mechanism. Therefore, we attribute the negative MR in our case to the excited-states-based channel MR(n).

In this context we can rule out the exciton–charge interaction model as an explanation for negative MR in TIPS-Pentacene FETs. This model predicts a magnetic-field induced increase in mobility. Since the mobility of the charge remains unchanged in an external magnetic field, the exciton–charge-interaction does not play a significant role in our device. Also the release of trapped charges due to an external magnetic field appears to be unlikely, because this process should not only be present under illumination but also in darkness. Therefore, this mechanism can also be ruled out in our case. Another explanation for negative MR in TIPS-Pentacene FETs that can be excluded is the bipolaron model. A magnetic field has no influence on the dark current and excited states have to be present to generate magnetic-field sensitivity. As conclusion, the bipolaron mechanism is not responsible for the photoinduced organic magnetoresistance in our devices. Finally, one possible explanation is the magnetic-field induced increase in *e-h* pair dissociation. The dissociation-reaction is dominated by weakly coulomb-correlated *e–h* pairs and the overall dissociation probability rises under the influence of an external magnetic field. Therefore more free charge carriers are generated, which results in (excited-states-based) negative MR.

4. Conclusion

We showed photoinduced negative MR of up to 0.57% in illuminated TIPS-Pentacene FETs measured at 60 mT and under irradiation intensity of 296 μW ($\lambda = 671 \pm 7$ nm). Thereby, the magnetic field exclusively influences the photocurrent of our device but not the dark current. The MR and photoresponse values are correlated and both are influenced by the gate voltage and do not depend on the drain voltage. We conclude that the observed photoinduced negative magnetoresistance in TIPS-Pentacene FETs is based on the presence of *e–h* pairs. The overall dissociation probability of *e–h* pairs rises under the influence of an external magnetic field, which leads to a higher number of free charge carriers and it results in negative MR. Finally, we want to point out that it is of high importance for answering fundamental questions about spin transport and spin manipulation in organic semiconductors, to develop specific MR models for organic field-effect transistors.

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