

# Phototuning Selectively Hole and Electron Transport in Optically Switchable Ambipolar Transistors

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One of the grand challenges in organic electronics is to develop multi-component materials wherein each component imparts a different and independently addressable property to the hybrid system. In this way, the combination of the pristine properties of each component is not only preserved but also combined with unprecedented properties emerging from the mutual interaction between the components. Here for the first time, that tri-component materials comprised of an ambipolar diketopyrrolopyrrole-based semiconducting polymer combined with two different photochromic diarylethene molecules possessing ad hoc energy levels can be used to develop organic field-effect transistors, in which the transport of both, holes and electrons, can be photo-modulated. A fully reversible light-switching process is demonstrated, with a light-controlled 100-fold modulation of *p*-type charge transport and a tenfold modulation of *n*-type charge transport. These findings pave the way for photo-tunable inverters and ultimately for completely re-addressable high-performance circuits comprising optical storage units and ambipolar field-effect transistors.

## 1. Introduction

Organic semiconductors are ideal active components for low-cost, flexible displays and large-area electronics, owing to their relative ease of processing from solution and at room temperature.<sup>[1]</sup> Ambipolar organic field-effect transistors (OFETs)<sup>[2]</sup> represent an advance towards the development of organic complementary metal oxide semiconductor (CMOS)<sup>[3]</sup> circuits by

incorporating both *p*- and *n*-type transistors. At the same time, blending semiconducting polymers with small molecules enables further tuning of optical and electrical properties, thus creating a powerful strategy to leverage the device performance.<sup>[4]</sup> Among the different functional molecules, photochromic molecules are capable of undergoing efficient and reversible photochemical isomerization between (meta)stable states, which exhibit markedly different properties such as light absorption, polarity, and redox behavior.<sup>[5]</sup> These molecules have been combined with organic semiconductors in order to control the overall material's properties and thus device performance with an optical input, resulting in light-responsive diodes,<sup>[6]</sup> transistors,<sup>[7]</sup> and memories.<sup>[8]</sup> Photochromic molecules were placed at the semiconductor–insulator interface,<sup>[9]</sup> as a self-assembled

monolayer on the electrodes,<sup>[10]</sup> or blended with organic semiconductors<sup>[11]</sup> to control optical and electrical characteristics. Among photochromic molecules, diarylethene derivatives<sup>[12]</sup> are very promising building blocks, combining good thermal stability and fatigue resistance.<sup>[13]</sup> These diarylethene moieties possess different highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels, depending on their stronger (closed isomer) or weaker (open

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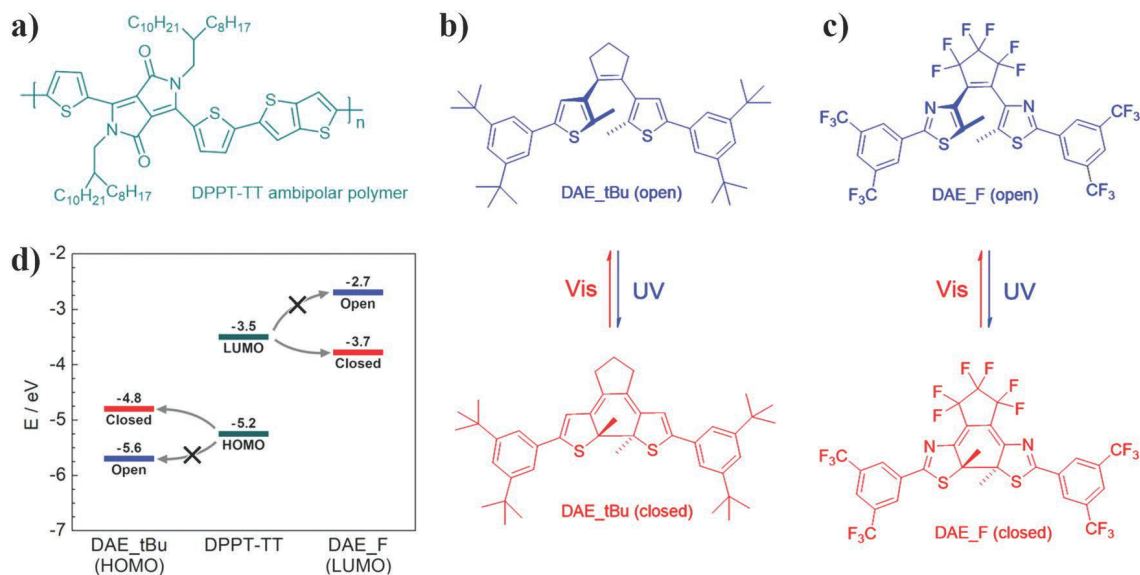
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**Figure 1.** Molecular structures and energy diagrams. Molecular structures of a) DPPT-TT, b) the open and closed form of DAE\_tBu, and c) DAE\_F. d) Schematic energy diagram representing the HOMO and LUMO levels of DPPT-TT ambipolar polymer and photochromic DAEs (DAE\_tBu and DAE\_F) in their open and closed states (energy levels measured by cyclic voltammetry).<sup>[15a,16,19]</sup>

isomer) extent of  $\pi$ -conjugation, which can be controlled upon irradiating at different wavelengths.<sup>[14]</sup> It was established that the incorporation of diarylethenes (DAEs) into either *p*-type<sup>[15]</sup> or *n*-type<sup>[16]</sup> semiconductors could ultimately lead to transistors with phototunable charge transport. The exceptional stability of these photochromic molecules in both open and closed forms could furthermore be exploited for the fabrication of non-volatile photoswitchable multilevel memories.<sup>[17]</sup> While phototunable energy levels could be introduced in *n*-type/DAE and *p*-type/DAE blends, the possibility to optically control electrons and holes *simultaneously* in an ambipolar semiconductor would be highly interesting towards fabricating light-controllable organic-based CMOS-like devices.

## 2. Results and Discussion

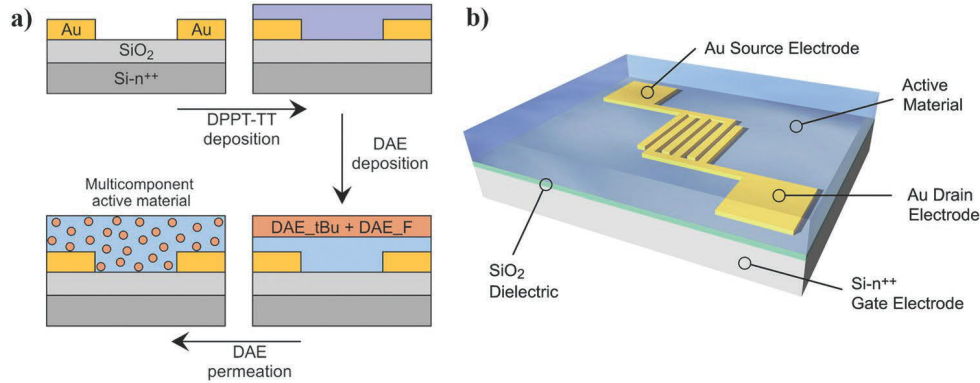
Here, we report on the fabrication and characterization of the first ambipolar para transistors whose *n*- and *p*-type currents can be selectively and reversibly photo-gated upon irradiation at different wavelengths. To this aim, we fabricate and characterize three-terminal devices whose active layer is based on simultaneous integration of two different DAEs derivatives into a polymer matrix capable of efficiently transporting both holes and electrons, demonstrating that the two photochromic units within the same polymer matrix retain their photo-responsive behavior upon blending.

Ambipolar transistors based on a diketopyrrolopyrrolethieno[3,2-b]thiophene copolymer (DPPT-TT, **Figure 1a**) as the active layer were fabricated in a bottom-contact, bottom-gate configuration, on a 230 nm SiO<sub>2</sub>/n<sup>++</sup>-Si substrate with pre-patterned gold electrodes ( $L = 20 \mu\text{m}$  and  $W = 10 \mu\text{m}$ ), by spin-coating of a 10 mg mL<sup>-1</sup> solution of DPPT-TT in dichlorobenzene. Two diarylethene derivatives highly robust to (optical) switching fatigue,<sup>[13]</sup> i.e., DAE\_tBu (**Figure 1b**) and DAE\_F (**Figure 1c**) dissolved in butyl acetate at 0.5 mg mL<sup>-1</sup>, were integrated into a

matrix of DPPT-TT. DPPT-TT was selected since it has previously been demonstrated<sup>[18]</sup> that it can be processed into ambipolar thin films featuring balanced hole and electron mobilities upon thermal annealing at 320 °C for 20 min. Given that the DAE derivatives are not stable at such a high temperature, these photochromic molecules were subsequently incorporated into the polymer matrix via temperature assisted permeation at a lower temperature, i.e., 90 °C (**Figure 2a**).

Owing to the electronic interactions between the chosen polymer and the DAE molecules, the output (drain) current of the as-fabricated OFET can be photo-tuned.<sup>[7,20]</sup> Depending on their isomeric form, i.e., open or closed switching state, DAEs feature different HOMO and LUMO levels (**Figure 1d**). The HOMO<sub>[DAE\_tBu, closed]</sub> and LUMO<sub>[DAE\_F, closed]</sub> lie at energy values, which are comprised within the DPPT-TT bandgap, and thus act as hole and electron accepting levels, respectively. In both cases, such mechanism results in a device current decrease. Importantly, the HOMO<sub>[DAE\_tBu, open]</sub> and LUMO<sub>[DAE\_F, open]</sub> levels do not fall within DPPT-TT bandgap, thus no opposing electronic interactions take place. In other words, hole and electron channels are clearly defined.

To investigate the effect of DAEs incorporation on the electron and hole mobilities, the ambipolar OFETs were characterized before and after adding DAEs into the polymer layer. All measurements were performed in the dark to avoid the undesired contribution of transiently photogenerated carriers. The electrical characteristics of OFET devices were measured in order to extract hole and electron field-effect mobilities ( $\mu_h$  and  $\mu_e$ ), threshold voltage ( $V_{th}$ ), and  $I_{on}/I_{off}$ . Device transfer curves recorded prior to DAE deposition (**Figure S3**, Supporting Information) displayed typical ambipolar characteristics with good hole transport characteristics ( $\mu_h \approx 0.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and  $I_{on}/I_{off} \approx 10^7$ ) but lower electron-transport characteristics ( $\mu_e \approx 0.012 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ,  $I_{on}/I_{off} \approx 10^5$ ). Mobilities could be further improved through octadecyltrichlorosilane (OTS) treatment of the dielectric support, reaching  $\mu_h \approx 0.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and  $\mu_e \approx 0.017 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ,



**Figure 2.** Device fabrication a) Schematic illustration of the preparation steps of OFET device in a bottom-contact bottom-gate geometry based on DPPT-TT polymer as the active layer with diarylethene molecules permeated into the matrix. b) Illustration of the final device.

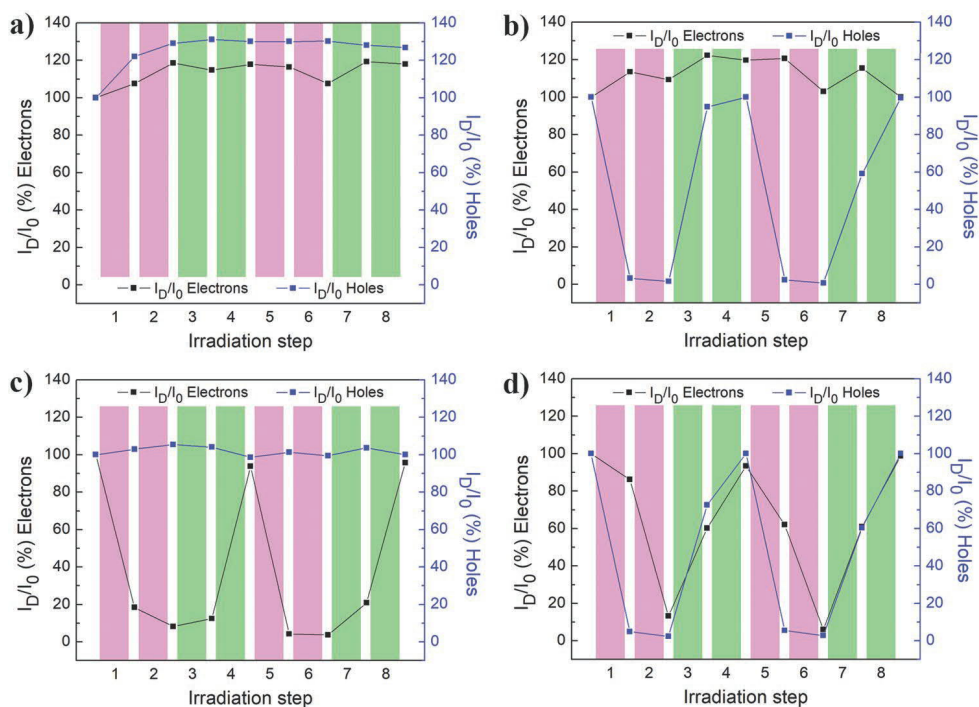
respectively. These mobility values were comparable with previous reports<sup>[21]</sup> on bottom-gate bottom-contact devices. Higher electron and hole mobilities ( $\mu_h \approx 1.36 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and  $\mu_e \approx 1.56 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) of the same polymer have been reported for top-gate bottom-contact devices.<sup>[18]</sup> To compare these performances, OFETs in top-gate bottom-contact configuration were fabricated (Figure S1, Supporting Information). The transfer characteristics for hole and electron transport as well as the electrical parameters are presented in the Supporting Information (Figure S2 and Table S1). These top-gate devices exhibited improved mobilities ( $\mu_e \approx 0.11 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ,  $I_{\text{on}}/I_{\text{off}} \approx 10^5$ , and  $\mu_h \approx 4.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ,  $I_{\text{on}}/I_{\text{off}} \approx 10^6$ ). However, since in a top-gate architecture most of the light during in situ irradiation would be absorbed by the gate metal and the insulating polymer, we focused our attention on DPPT-TT-DAE transistors in bottom-gate bottom-contact configuration.

Bi- and three-component devices based on DPPT-TT with 10 wt% of one or two DAE molecules (DAE\_tBu and DAE\_F) as the active layer were fabricated. Comparison of the electrical performances of OFETs before and after DAEs deposition (Figure S3, Supporting Information) revealed that DAEs incorporation resulted in a small decrease in mobility (Table S2, Supporting Information). The morphology of pristine polymer and DAE-containing films was studied by atomic force microscopy (AFM) by imaging single, bi- and three-component devices (Figure S4, Supporting Information). The AFM analysis revealed unaltered film morphology on the micrometer scale upon permeation of DAEs through the polymer film, characterized by tightly packed lamellar structures. The root mean square roughness ( $R_{\text{RMS}}$ ) of the different films was determined in an area of  $5 \times 5 \text{ }\mu\text{m}^2$  (Table S3, Supporting Information) and displays only slightly higher values in bi- and three-component-based devices ( $R_{\text{RMS}} = 1.47\text{--}1.79 \text{ nm}$ ) when compared to pristine DPPT-TT polymer films ( $R_{\text{RMS}} = 1.14 \text{ nm}$ ). To cast further light onto the effect of DAEs incorporation on the crystalline structure of the polymer, grazing incident X-ray diffraction measurements were performed (Figure S10 and Table S4, Supporting Information). These measurements revealed that the permeation of DAEs into the polymer matrix did not result in any major disruption of semiconductor molecular packing. Such result is in accordance with previous reports of DAEs incorporated in a polymer matrix.<sup>[15b]</sup>

The effect of light irradiation wavelength on the drain current was investigated by exposing bi- and tri-component OFETs to ultraviolet ( $\lambda = 365 \text{ nm}$ ) and green ( $\lambda = 546 \text{ nm}$ ) LEDs from the top of the sample surface under nitrogen atmosphere. Light irradiation was found to induce a marked modulation of hole and electron drain current as shown in **Figure 3** for devices of DPPT-TT with permeated DAE\_tBu (Figure 3b), and with permeated DAE\_F (Figure 3c). Remarkably, it was possible to induce both hole and electron drain current modulation in the three-component devices (Figure 3d). As expected, devices based on the pristine polymer DPPT-TT did not exhibit any change in current upon irradiation (Figure 3a).

In the bi-component devices based on the polymer with inserted DAE\_tBu molecules (Figure 3b), the hole drain current decreased by two orders of magnitude upon UV illumination. Upon irradiation with visible light, the current recovered to 98% of the initial value, while the electron current remained constant upon irradiation in the UV and in the visible. In DPPT-TT/DAE\_F OFETs, the electron current was modulated upon UV and visible illumination by a factor of 10, while the hole current remained stable. Upon UV light irradiation, the drain current extracted from three-component devices exhibited a decrease of 92% and 85% for holes and electrons, respectively (Figure 3d). After visible light irradiation (two steps of 5 min), the drain current of the electron and hole increased and completely recovered. The ability to optically switch the output current of our OFETs indicates that DAE molecules efficiently diffuse through the polymer matrix and reach the semiconductor-dielectric interface.

These results confirm the light-switching process, which is completely reversible and selective for both hole and electron charge transport. Furthermore, the large light-induced current modulation was maintained, even after the devices were stored for several months inside the glove box, indicating that small diarylethene molecules non-covalently bound into polymer matrices can be used in robust optical memories. While the device still operated properly with similar mobility (around  $0.03 \text{ cm}^2 \text{ Vs}^{-1}$ ) upon 18 months of storage in the glovebox (in the dark), a shift in threshold voltage (from  $-17$  to  $-3 \text{ V}$ ) was observed as the result of the exposure to a finite amount of oxygen, which is present in ca. 50 ppm in our glove box.



**Figure 3.** OFET characteristics under ultraviolet and visible light irradiation.  $I_D/I_0$  of OFET devices based on a) pristine DPPT-TT, b) DPPT-TT with DAE\_tBu (10 wt %), c) DPPT-TT with DAE\_F (10 wt %), and d) DPPT-TT with both 10 wt % of DAE\_tBu and 10 wt % of DAE\_F. Violet and green shaded areas correspond to the irradiation with light at wavelengths of 365 and 546 nm, respectively. The irradiation is performed from above the devices. All irradiation steps power and duration are detailed in Section 1 of the Supporting Information.

The retention of the photoswitching ability of DAEs diffused into DPPT-TT matrix was further investigated by UV/visible absorption spectroscopy. UV and visible light irradiation on neat DAE\_tBu and DAE\_F in solution (Figure S6, Supporting Information) and in films (Figure S9, Supporting Information), resulted in the appearance and disappearance of the absorption band between 400 and 600 nm. The photocyclization and photocycloreversion of both DAE\_tBu and DAE\_F have been previously reported, highlighting good reliability over multiple cycles of alternating UV and visible light irradiation.<sup>[15a,16]</sup> After diffusion into the DPPT-TT film, UV irradiation at  $\lambda = 312$  nm led to an increase in absorption in the  $\lambda = 400$ –600 nm region for both DAE\_tBu (Figure S7a, Supporting Information), and DAE\_F films (Figure S8a, Supporting Information), corresponding to the formation of the closed-ring isomer.<sup>[22]</sup> Subsequent irradiation of the films with visible light ( $\lambda = 530$  nm), to switch the molecules back to the open-ring isomer, resulted in spectral recovery of the initial states (Figures S7b for DAE\_tBu and S8b for DAE\_F in the Supporting Information), confirming the reversible nature of DAE photo-switching in the DPPT-TT films.

The photochemical quantum yield (QY) for the photoisomerization from the ring-closed form to the open-ring isomer of DAEs in the polymer matrix was determined as  $\Phi = 0.003$  for DAE\_tBu and  $\Phi = 0.02$  for DAE\_F. The lower QYs, compared to those measured in solution ( $\Phi = 0.009$  for DAE\_tBu and  $\Phi = 0.039$  for DAE\_F),<sup>[13,15a]</sup> are expected as the polymer matrix hinders the change in the molecular configuration. Nevertheless, these QYs remain high enough to enable an efficient photoswitching of the output current in the OFETs.

### 3. Conclusion

In summary, photomodulation of the output current in ambipolar field-effect transistors was achieved through the fabrication of multicomponent devices based on an active layer of DPPT-TT permeated with DAEs. The light-switching process in films was completely reversible and selective using DAE\_tBu and/or DAE\_F as photochromic molecules. While previous reports have relied on bi-component designs (a single semiconductor with a single photochromic unit), the tri-component transistors fabricated here offer a clear demonstration that the device functional complexity can be increased by the addition of multiple different photochromic units with complementary functions within the polymer matrix.

The drain current of the OFETs was reversibly modulated by alternating ultraviolet and visible light irradiation, with a 100- and 10-fold modulation of charge transport in the *p*- and *n*-channels, respectively. Most importantly, extreme versatility was demonstrated as one and the same polymer, DPPT-TT, was used and processed in the identical fashion to produce three types of photoswitchable OFETs (*p*-type, *n*-type, and ambipolar) upon selected diarylethene diffusion. This finding is of paramount practical relevance, as it demonstrates that large-area electronics could be easily produced from a homogeneous high-performance ambipolar film, and selectively modified to obtain ambipolar, *p*- and *n*-type conducting as well as reduced-conductivity regions through careful selection, deposition, and operation of photoswitchable moieties. Since the diffusion method demonstrated no adverse effect on the charge transport (both for *p*- and *n*-type carriers), this approach can be

employed without loss of performance, unlike the previously reported blending method, that resulted in a twofold decrease in current as compared to the semiconductor in the absence of added photochromic units.<sup>[17]</sup> This is of crucial importance to develop future optical memories with higher current values for improved readout accuracy. The modulation of the ambipolar charge transport with conservation of high currents when the photochromic units are in the open form marks a first step in the fabrication of inverter circuits with a photomodulable gain once a semiconducting polymer with a balanced ambipolarity is chosen. While this specific system relies on two DAEs switching at the same wavelength, this system paves the way for the future development of smart materials with independently addressable photochromic units. This could result in photoswitchable ambipolar transistors with separate hole and electron charge transport modulation, leading to circuits that could be completely redesigned by simple light-based writing for a new generation of evolutive electronics.

## 4. Experimental Section

Experimental details are reported in the Supporting Information.

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## Conflict of Interest

The authors declare no conflict of interest.

## Keywords

ambipolar transport, diarylethenes, organic field-effect transistors, photochromic molecules, semiconducting polymers

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