

## Review

# Photoresponsive organic field-effect transistors involving photochromic molecules



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## ABSTRACT

In recent years, organic field-effect transistors (OFETs) with high performance and novel multi-functionalities have attracted considerable attention. Meanwhile, featured with reversible photo-isomerization and the corresponding variation in color, chemical/physical properties, photochromic molecules have been applied in sensors, photo-switches and memories. Incorporation of photochromic molecules to blend in the device functional layers or to modify the interfaces of OFETs is common way to build photo-transistors. In this review, we focus on the recent advantages on the study of photoresponsive transistors involving one of three typical photochromic compounds spiropyran, diarylethene and azobenzene. Three main strategies are demonstrated in detail. Firstly, photochromic molecules are doped in active layers or combined with semiconductor structure thus forming photo-reversible active layers. Secondly, the modification of dielectric layer/active layer interface is mainly carried out by bilayer dielectric. Thirdly, the photo-isomerization of self-assembled monolayer (SAM) on the electrode/active layer interface can reversibly modulate the work functions and charge injection barrier, result in bifunctional OFETs. All in all, the combination of photochromic molecules and OFETs is an efficient way for the fabrication of organic photoelectric devices. Photoresponsive transistors consisted of photochromic molecules are potential candidate for real applications in the future.

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## 1. Introduction

Organic field-effect transistors (OFETs), one of the most significant branches of modern organic electronics, have earned keen interest in recent decades for their potential applications in flexible large-area display, smart tags and cards, and physical/chemical sensors [1–4]. On the one hand, as more and more innovative  $\pi$ -conjugation organic semiconductors are designed and synthesized, carrier mobility, the major electrical characteristic of OFET, has surpassed that of amorphous silicon ( $0.1\text{--}1.0\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ ) and reached  $5\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$  or even higher for both small molecules and polymer, p-type and n-type organic semiconductors [5–13]. In addition, solution-processed OFETs with high performance (high charge mobility, low threshold voltage and high current on/off ratio) and stability have experienced remarkable improvements [14–17].

However, the commercial application of OFETs is still limited due to the relatively lower device performance, not ideal device stability and the challenging device integration technology. On the other hand, great efforts have been devoted to the design and fabrication of multi-functional OFETs. As a result, combining traditional OFETs with functional molecular building blocks to install multi-functional OFETs has been the latest research tendency. Considerable novel multi-functional devices based on OFET such as phototransistors [18–20], light-emitting transistors [21,22], sensors [23,24] and memories [25,26], have been achieved by molecular tailoring, modification of dielectric layer and electrode layer, construction of active layer, etc. Some reviews on device fabrication strategies and challenges have been previously published [27,28]. Smart use of these organic electronic devices by the introduction of stimuli responsive materials is one of the common ways [29–32]. Organic functional materials that are sensitive to gas, light, pressure, temperature and pH have already been employed to OFETs, affording multifunctional devices such as sensors and switches [24,33–38]. Phototransistors particularly the devices that are

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electrically and optically responsive due to the integration with photochromic molecules are attracting more and more attention [39–42]. Photochromic molecules can reversibly switch between two isomers result in the reversible color change simultaneously, when exposed to particular wavelength of light. Nonionic-ionic spiropyran, open ring-closed ring diarylethene and *trans-cis* azobenzene are three main sorts of photochromic systems frequently used in photoresponsive OFETs [43]. Bi-functional OFETs combined with these photo-switchable molecules are promising candidates for new applications in light sensors and optical memory where the photostimulation can be precise controlled on direction, intensity and wavelength [44].

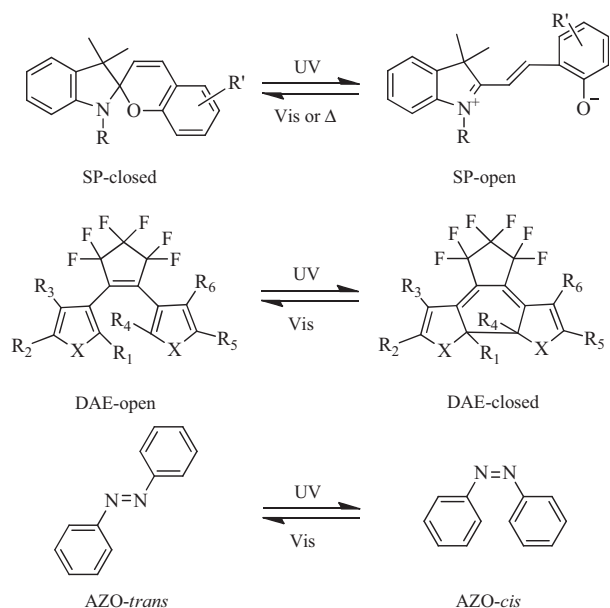
A typical OFET is composed of an active layer, a dielectric layer, a gate electrode and source/drain electrode as well as their corresponding interfaces including electrode/active layer interface, dielectric/active layer interface, and organic/organic layer interface [45]. Both the functional layers and interfaces have key influences on the device performance. In this contribution, we attempt to present a brief overview on recent advances in the study of photoactive OFETs involving photochromic molecules in active layers, in dielectric layers, and on the electrode/active layer interface.

## 2. Photochromic molecules

Photochromic molecules can reversibly switch between two isomers induced by light or heat, where the two forms have different absorption spectra and cause the discoloration intuitively. Generally, the variation from colorless isomer to colored isomer is induced by ultraviolet (UV) irradiation, and the restoring course to the initial state can be performed by visible (Vis) light or thermal relaxation. During the isomerization, the physical and chemical properties of these photochromic compounds can also be changed, including refractive indices, dielectric constants, geometrical structures, energy levels, polarity, magnetic properties and oxidation/reduction potentials [43,46]. These controllable property tunings have inspired people to apply photochromic system in photoelectric field such as photo-switches [47–49], sensors [50,51], molecular machine [52–54], data storage [55–57], liquid crystal [58–60], and so on. Spiropyran, diarylethene and azobenzene are three most widely studied organic photochromic molecules, and their structures and the photochromism process of these molecules are presented in Fig. 1.

Spiropyran (SP) is one of the oldest families of photochromic molecules. As shown in Fig. 1, the cleavage of the C<sub>spiro</sub>-O bond in the parent colorless SP-isomer was triggered by UV light, making the spiro structure transform into the merocyanine form. The recovery process can easily occur either by exposure to visible light or by thermal relaxation. Besides the color variance, the open and closed SP forms differ a lot in electric dipole moment, structure, emission behavior, protonation and chemical affinity, thus make the SP responsive to multiple stimuli such as light, pH [61], metal ions [62,63] and heat. Based on this unique characteristic, many investigations on the study of SP-based dynamic materials have been reported such as SP-functional polymers [64], light-controlled surface wettability [65], photo-control inorganic nanoparticles and so on [66].

It is generally accepted that diarylethene (DAE) derivatives bearing two thiophene-derived groups is the most suitable photochromic compounds for practical application. The most outstanding features of DAE compounds are their relatively high light-sensitivity, fatigue resistance and thermal stability [46,67]. Photochromism mechanism of DAE is shown in Fig. 1. With the irradiation of UV/vis light, DAE switches between an open-ring form and closed-ring form. Highlighted for the thermal stability of both isomers, DAE molecules have become the hottest system for



**Fig. 1.** Photochromic mechanism of three typical photochromic molecules. top, spiropyran from SP-closed to SP-open; middle, diarylethene from an open-ring form to a closed-ring form; bottom, azobenzene from a *trans*-isomer to a *cis*-isomer.

light-triggered photo-switches, like fluorescent switch, supramolecular switch, liquid crystal switch, crystalline and amorphous switch. These DAE-based photo-switches have been summarized in Tian's excellent review [49].

Discovered in the mid-18th century, azobenzene (AZO) was once regarded as a coloring agent used for the synthesis of desired dyes until its *cis*-form was firstly found by Hartley in 1937 [68]. Composed of two phenyl rings linked by an azo linkage (-N=N-), AZO can switch between two *cis-trans* isomers induced by particular wavelengths of light. It is demonstrated that the *cis* form can convert back to the *trans* isomer either by vis light or by thermal relaxation in the dark. This steady transformation rate depends on the light intensity and wavelength, ambient temperature, and the host matrix (be in solution, liquid crystal, monolayer, or polymer) [69]. A large number of expending researches on photobiology [70,71] and photoelectrochemical information storage [59,72–74] have emerged according to the facile geometric isomerization of AZO.

## 3. Photochromic molecule in OFET active layer

The performance of OFETs mainly depend on the properties of the device active layer where free charge carriers transport between the source and drain electrodes and come into being of source/drain current ( $I_{DS}$ ) under a gate voltage. Bifunctional OFETs with photochromic molecules and organic semiconductors are modulated by light and voltage simultaneously. The most direct way to realize this attempt is by blending photochromic molecules with organic semiconductors in the active layer. In this way, different photochromic isomers would have different doping effects on the organic semiconductors and reversibly modulate the output signal.

### 3.1. Photochromic molecule doped OFET active layer

#### 3.1.1. Diarylethene doped OFET active layer

As mentioned above, diarylethene (DAE) is a potential photochromic system for photo-triggered OFETs thanks to its thermal and energy levels stabilities in both isomers. In 2012, Samori et al. [39] fabricated bi-component OFETs based on DAE

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