

Photo-induced negative differential resistance of organic thin film transistors using anthracene derivatives

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ABSTRACT

We fabricated organic thin film transistors (OTFTs) using soluble 5,5'-(2,6-Bis((4-hexylphenyl)ethynyl)anthracene-9,10-diyl)bis(ethyne-2,1-diyl)bis(2-hexylthieno[3,2-*b*]thiophene (HTT-ant-THB) as an active layer. We studied the photo-responsive and the gate field-dependent charge transport characteristics of the HTT-ant-THB-based OTFTs. When light ($\lambda_{\text{ex}} = 505 \text{ nm}$) was irradiated on the OTFTs, negative differential resistance (NDR) behavior (i.e., negative slope of the current versus voltage curve) was observed in the reverse bias region of the source-drain current versus voltage characteristics. The NDR effect observed in this study is unique and is controlled by the wavelength and power of the incident light. The current hysteresis and NDR characteristics can be explained in terms of the trapping and releasing mechanism of the mobile charges at the interface between the electrodes and the organic layer. In addition, the NDR effect in the device disappeared on applying negative gate bias.

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1. Introduction

Small molecule organic semiconductors with a π -conjugated structure show promising electrical and optical properties for optoelectronic devices, such as a relatively high charge carrier mobility and a tunable optical absorption [1,2]. The functionalized organic small molecules exhibit good solubility in organic solvents, enabling flexible thin film processes at relatively low temperatures for organic thin film transistors (OTFTs), organic photovoltaic cells (OPVCs), and organic-based sensors [3–5]. The devices using organic small molecules with the relatively high photoluminescence quantum yield (PLQY) [6] have also shown photo-responsive charge transport characteristics because of the generation of photo-induced charges

and their dissociation into free mobile charge carriers, resulting in the photovoltaic and/or memory effect. Narayan and Kumar [7] reported on the photo-responsive OTFTs using poly(3-octylthiophene-2,5-diyl). They also studied the optoelectronic memory elements for the photo-responsive OTFTs using poly(3-hexylthiophene) [8]. Organic photo-transistors (OPTs) using soluble and star-shaped oligothiophenes with four-armed π -conjugations as an active layer were also reported [9]. Recently, Pyo et al. [10] reported the photosensitive organic complementary inverter.

The negative differential resistance (NDR) can be characterized by a negative slope of the source-drain current versus voltage ($I_{\text{ds}}-V_{\text{ds}}$) characteristic curve. The light-induced NDR effect of the inorganic $p-i-n$ photo-diodes using InP/InGaAs/InP was reported by Wu et al. [11]. They analyzed the light-induced NDR effect in terms of the transferred electron effect. The NDR was caused by the average velocity drop as an amount of electrons to be accumulated near the potential barrier of InGaAs. A second

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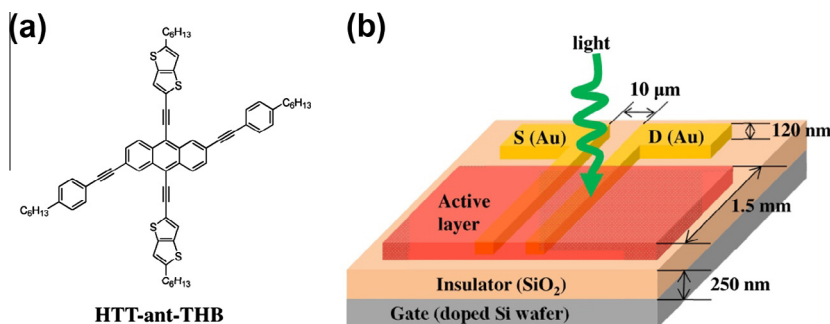
photocurrent drop occurred in the depleted InP region. The photon-assisted tunneling occurring around the InGaAs *pn* junction, which causes additional absorption, affects the drop in the photocurrent. The NDR phenomenon has been utilized in organic-based nonvolatile resistive memory devices [12]. The structural and electrical anisotropy of organic materials induces an anisotropic charge conduction and the electrical hysteresis curves [13]. Because of the switching capability using two different resistance states and hysteresis, some of the π -conjugated polymers and small molecules were used for active layers in the memory devices [14,15]. The NDR effect and the switching mechanism of the organic-based memory devices can be explained in terms of filament mechanism, charge trapping/de-trapping, and/or formation of an oxide layer on interface, etc. [16–18]. Light-induced NDR effects in the organic small molecule-based OTFTs have not yet been reported.

In this study, we fabricated the OTFTs using the soluble 5,5'-(2,6-Bis((4-hexylphenyl)ethynyl)anthracene-9,10-diyl)bis(ethyne-2,1-diyl)bis(2-hexylthieno [3,2-b]thiophene (HTT-ant-THB) as an active layer. We observed the NDR and hysteresis characteristics from the photo-responsive *I*–*V* characteristic curves of the devices when light was irradiated on them. The NDR effect could be explained by the trapping and de-trapping mechanism of photo-induced charges at the interface. We also observed that the NDR effects were controlled by the gate bias and the incident light power and wavelength. The tunable NDR effect of the OTFTs can be applied to sensors and memory devices in an atmospheric condition.

2. Experimental

2.1. Fabrication of OTFTs using HTT-ant-THB

Schematic 1a shows the molecular structure of HTT-ant-THB. The synthesis of HTT-ant-THB molecules was previously reported by Jo et al. [19]. Ten milligrams of HTT-ant-THB was dispersed in 1 ml of chloroform (CHCl₃) solution using an ultrasonic cleaner at 60 °C for 90 min. Schematic 1b shows the device structure of the HTT-ant-THB-based OTFT. The doped *p*-type Si wafer ($R = 0.001\text{--}0.003 \Omega \text{ cm}$) and the thermally grown SiO₂ layer were used as the gate electrode and the dielectric layer, respectively. The thickness and dielectric constant (ϵ_r) of the SiO₂ layer were $\sim 250 \text{ nm}$ and ~ 3.9 , respectively.



Scheme 1. (a) The schematic chemical structure of the HTT-ant-THB molecule. (b) The schematic device structure of the HTT-ant-THB-based OTFT.

Using conventional photolithography, gold (Au) source and drain electrodes were patterned with a length and width of 5–10 μm and 1500 μm , respectively. For better adhesion, a Ti layer of $\sim 5 \text{ nm}$ thickness was deposited before the deposition of the Au electrodes. The devices were treated with ozone for 10 min and then spin-coated with hexamethyldisilazane (HMDS) at 4500 rpm for 30 s for a better match between the organic layer and the SiO₂ layer. The HTT-ant-THB solution was then spin-coated on the HMDS treated SiO₂ layer at 2000 rpm for 30 s for the formation of active layer.

2.2. Measurements and characterizations

Collimated light emitting diodes (ZEISS Axioskop, Thorlabs Inc.) were used as the light source. The wavelengths and powers of the incident light were controlled in the range of 505–735 nm and 1.05–4.82 mW cm^{-2} , respectively. The distance between the light source and device was fixed at 3.5 cm. More than 100 OTFT devices were fabricated to measure the photo-responsive and gate field-dependent *I*–*V* characteristic curves. Keithly 237 SMU was used as the source and measuring instrument of current and/or voltage under atmospheric or vacuum conditions at room temperature (RT). We used a rotary pump for the vacuum condition (under 25 m Torr). The surface morphology of the HTT-ant-THB thin films was measured by an atomic force microscope (AFM, Nano-Focus Ltd., Albatross). The scan size and rate were $10 \times 10 \mu\text{m}^2$ and 0.5 Hz, respectively.

3. Results and discussion

3.1. Hysteresis curves of the HTT-ant-THB-based OTFTs

Fig. 1a and its inset show the I_{ds} hysteresis curves as a function of V_{ds} of the HTT-ant-THB-based OTFTs ($V_{\text{g}} = 0 \text{ V}$) under light and dark states, respectively, in an atmospheric condition. The $I_{\text{ds}}\text{--}V_{\text{ds}}$ characteristic curves ($V_{\text{g}} = 0 \text{ V}$) of the devices show an asymmetric behavior, such as a rectification effect. In the dark state, the hysteresis was observed only in the forward bias region and not in the reverse bias region. The asymmetric $I_{\text{ds}}\text{--}V_{\text{ds}}$ behavior might be due to the difference in the Schottky barrier height and width in the two contact regions of the metal electrodes and the semiconductor and/or the asymmetric impurity driven

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