Organic Electronics 15 (2014) 2486–2491

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Photon-energy-dependent light effects in organic nano-floating-gate nonvolatile memories

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ARTICLE INFO

Article history:
Received 17 May 2014
Received in revised form 5 July 2014
Accepted 13 July 2014
Available online 29 July 2014

Keywords:
Organic field-effect transistors
Nonvolatile memory
Illumination
Pentacene

ABSTRACT

A pentacene-based organic field-effect transistor nonvolatile memory, in which polystyrene covered Au nanoparticles act as the nano-floating-gate, is probed under different illumination conditions. The memory window can be greatly enlarged upon illumination depending on incident photon energy and intensity, and two light effects are proposed and discussed. The minority multiplication effect enhances the minority carrier tunneling into the nano-floating-gate, resulting in the remarkable positive V_T shift. The excitation-induced injection effect is strongly photon energy dependent, and it is responsible for the significant negative V_T shift. Appropriate illumination is favorable for reducing the programming/erasing voltage of organic nano-floating-gate nonvolatile memories.

1. Introduction

Development of organic nonvolatile memories promotes the applications of flexible electronics requiring integration of data processing and storage [1,2]. Owing to the advantages of large memory capacity, nondestructive readout, and architectural compatibility with integrated circuits composed of organic field-effect transistors (OFETs), organic nonvolatile memories based on nano-floating-gate have been intensively studied [3–10]. Typical nano-floating-gate such as metal nanoparticles (NPs) offers high-density and spatially discrete charge trapping sites, which are favorable for suppressing charge leakage from floating-gate and scaling down tunneling dielectric [8–10]. In contrast to conventional Si-based FETs which can work in either the accumulation or inversion mode, OFETs normally operate in the accumulation mode due to unipolar behavior of most organic semiconductors. The deficiency of minority carriers in organic active layers will impede the charge trapping into nano-floating-gate during programming [11–13], resulting in a severe reduction in memory window. The number of minority carriers can be effectively increased by appropriate optical excitation, and thus light-assisted programming and/or erasing have been utilized to facilitate the charge trapping/detrapping process in OFET-based memories [8,12,14–16]. However, the fundamental of the light effects, especially the photon energy dependence of memory performance, needs to be clarified with a clear physical picture. It could allow an in-depth probing of the charge trapping mechanism in organic nano-floating-gate nonvolatile memories.

In this report, the light responses under different illumination conditions of an OFET-based memory were systematically probed, where pentacene, Au NPs and polystyrene (PS) are employed as the organic active layer, nano-floating-gate and tunneling dielectric, respectively. The memory window is strongly dependent on incident photon energy and intensity, arising from the minority multiplication effect and/or excitation-induced injection effect. Both the light effects are interpreted by a physical picture of the photon-energy-dependent charge trapping from pentacene

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http://dx.doi.org/10.1016/j.orgel.2014.07.018
1566-1199/

into the nano-floating-gate, where both electron and hole trapping occur but behave distinctly. It is demonstrated that illumination can largely reduce the programming/erasing voltage in the organic nano-floating-gate memory.

2. Experimental

2.1. Device fabrication

The memory device structure is illustrated in Fig. 1. A heavily-doped Si wafer with 200-nm-thick SiO_2 on top was used as the control-gate covered with the control dielectric. After routine substrate cleaning, 3-nm-thick Au was deposited on the SiO_2 surface by simple sputtering with a desktop sputter system (Quorum Technologies) to spontaneously form the Au NPs. Subsequently, a PS thin layer of about 11 nm acting as the tunneling dielectric was deposited by spin-coating, followed by annealing in a glove box at 100 °C for 15 m [9]. Then 40-nm-thick pentacene (Tokyo Chemical Industry, purified by vacuum sublimation before using) was deposited on the PS surface by vacuum deposition (Kurt J. Lesker, working pressure <10^{-6} Torr), where the deposition rate and substrate temperature were kept at 0.02 nm/s and room temperature, respectively. In the final step, Cu drain/source electrodes were prepared by thermal evaporation through a sophisticated shadow mask [17], by which the conducting channel length (L) and width (W) are defined as 100 μm and 750 μm, respectively.

2.2. Device characterization

The film thicknesses were estimated with atomic force microscopy (AFM, Veeco) and confirmed with an ellipsometer (J.A. Woollam). The electrical characteristics of the organic nano-floating-gate memory were measured at room temperature using a semiconductor parameter analyzer (Keithley 4200) in a high-vacuum probe station (Lake Shore, base pressure <10^{-5} Torr). For the light response measurements, laser pointers with different wavelengths of 408 nm (Blue–Violet (B–V), 3.0 eV), 532 nm (Green, 2.3 eV) and 652 nm (Red, 1.9 eV) were used as the light sources, whose luminous spectra recorded by a spectroradiometer (Photo Research) are shown in Fig. 1. The full width at half maximum (FWHM) of all the spectra is about 9 nm. The light intensity was modulated by neutral-density filters, and measured with an optical power meter precisely at the device position.

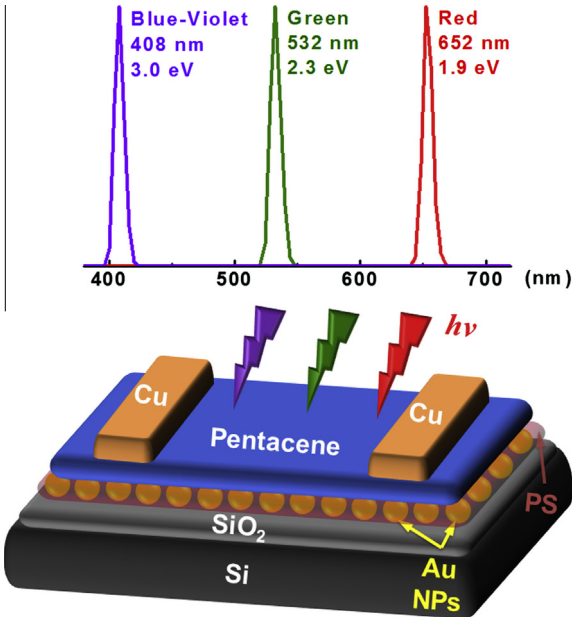

Fig. 1. Scheme of a pentacene-based OFET nonvolatile memory under illumination, where Au NPs and PS layer are employed as nano-floating-gate and tunneling dielectric, respectively. Three light sources are used for probing the light effects on memory performance, whose luminous spectra are shown on top.

3. Results and discussion

Fig. 2a, c and e shows the transfer characteristics at the drain biases (V_{DS}) = −3 V of the organic nano-floating-gate memory measured in dark and under different illumination conditions. Both the positive and negative threshold voltage (V_T) shifts, corresponding to electron and hole trapping into the nano-floating-gate at high positive and negative gate biases (V_{GS}), respectively [3,4,9,13], take place to generate a large memory window. The memory window could be further enlarged by increasing the Au NP size, as reported by Han et al. [7]. The FE mobility (μ_{FE}) of the device is about 0.8 cm^2/Vs regardless of illumination conditions. Furthermore, the memory shows good retention capability at both ON and OFF states no matter which illumination condition was executed, as demonstrated in Fig. 2b, d and f. By comparing the device data measured in dark before and just after illumination, the coincidence of the transfer characteristics demonstrates that light-induced heating effect on the drain current (I_D) can be neglected (see Fig. S1 in Supplementary material), and other optical excitation effects should be taken into account.

For the red light (Fig. 2a), the positive V_T shift increases upon raising the light intensity, while the negative V_T shift is not changed. It indicates that electron trapping into the nano-floating-gate is sensitive to the red light absorption but hole trapping is not [11–13]. The phenomenon appears more prominent using the green light (Fig. 2c), and a larger memory window is generated due to the greater positive V_T shift. Pentacene is a typical p-type organic semiconductor, and the energy gap between its highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) is about 1.9 eV [18,19], which is similar to the photon energy of the red light. As the charge trapping process in the organic nano-floating-gate memory is considered to be dominated by the Fowler–Nordheim tunneling mechanism [4,20], the deficient electron (minority carrier) supply in LUMO of pentacene at high positive V_{GS} (programming) may limit the positive V_T shift [11–13]. The employment of illumination during programming can largely increase the minority carrier density [21,22], which will enhance electron trapping into the nano-floating-gate and result in an increase in the positive V_T shift. This minority multiplication effect is reinforced when the