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Interfacial dipole in organic p–n junction to realize write-once-read-many-times memory





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ABSTRACT

A new approach is exploited to realize nonvolatile organic write-once-read-many-times (WORM) memory based on copper phthalocyanine (CuPc)/hexadecafluoro-copper-phthalocyanine (F_{16} CuPc) p-n junction. The as-fabricated device is found to be at its ON state and can be programmed irreversibly to the OFF state by applying a negative bias. The WORM device exhibits a high ON/OFF current ratio of up to 2.6×10^4 . An interfacial dipole layer is testified to be formed and destructed at the p-n junction interface for the ON and OFF states, respectively. The ON state at positive voltage region is attributed to the efficient hole and electron injection from the respective electrodes and then recombination at the CuPc/F₁₆CuPc interface, and the transition of the device to the OFF state results from the restricts charge carrier recombination at the interface.

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1. Introduction

In recent years, organic resistive memory devices in which the active organic materials process at least two stable resistance states have been extensively investigated due to their simplicity in device structure, good scalability, low-cost potential, low-power operation, and large capacity for data storage [1–15]. There are three types of organic memory devices: random access memory, read-write-erase-rewritable memory, and write-once-read-many-times (WORM) memory. WORM memory is a type of nonvolatile memory that is capable of storing data permanently and being read from repeatedly. Thus it bears the potential application for permanent data storage, such as wireless identification tags, smart cards, and personal data depositories. A wide variety of materials, such as small molecular organic material [16,17], polymer [18–20],

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organic/inorganic heterojunction [21,22], conjugated copolymer [23], organic donor/acceptor composite [24], have been explored for organic WORM memory. Correspondingly, the mechanisms accounting for the transition between the high conductivity (ON) and low conductivity (OFF) states include charge carrier traping and detraping [16–18], filament formation and destruction [19], conformation change [20], oxidation/reduction reaction [21,22], charge-transfer complex formation [23,24], and etc.

Organic p–n junction has attracted much interest in relation to the rapid development of the organic optoelectronic devices, such as organic light-emitting diodes [25], solar cells [26], photodetectors [27], and field-effect transistors [28]. Band bending and interfacial dipole are often found in organic/organic p–n junctions, and such effects play crucial roles in determining the performance of the multilayer organic optoelectronic device [29–32]. Copper phthalocyanine (CuPc) and hexadecafluoro-copper-phthalocyanine (F_{16} CuPc) are hole and electron transporting materials, respectively, with high field-effect mobilities both in the order of 10^{-2} cm²/(V s) as well as excellent thermal stability [33,34]. Lin and Ma have observed WORM



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memory characteristic in CuPc by controlling the evaporation rate and the ON/OFF transition was proposed to be managed by charge carrier traping and detraping [17]. Choi et al. have observed WORM memory characteristics based on a hyperbranched CuPc polymer, and the ON/OFF transition was proved to be governed by the rupture of filaments [19]. In a previous work, we have demonstrated an organic WORM memory in device indium tin oxide (ITO)/F₁₆CuPc/ Al with an ON/OFF current ratio of 2.3×10^3 , and the ON/ OFF transition results from the formation and destruction of the interfacial dipole layer formatted in the ITO/F₁₆CuPc interface [35].

In this study, a nonvolatile organic WORM memory device based on $CuPc/F_{16}CuPc p-n$ junction is demonstrated. The device shows an ON/OFF current ratio in the order of 10^4 . And a new mechanism is proposed to govern the conductivity transition. The ON state of the WORM device is attributed to the low barrier for charge carriers injection and then recombination at the $CuPc/F_{16}CuPc p-n$ junction interface. The transition from the ON state to the OFF state by applying a negative bias results from the destruction of the interfacial dipole layer which forms an insulating layer and hence restricts charge carriers recombination in organic layers.

2. Experimental details

Devices were fabricated on patterned ITO coated glass substrates with a sheet resistance of 15 Ω /sq. The substrates were routinely cleaned and treated in an ultraviolet–ozone environment for 10 min before loading into a high vacuum chamber. Organic layers and Al cathode were deposited onto the substrates via thermal evaporation at 5×10^{-4} Pa. Two devices with the configurations were fabricated as follows:

- Device A: ITO/CuPc (40 nm)/F₁₆CuPc (40 nm)/Al (100 nm)
- Device B: ITO/CuPc (30 nm)/4,7-diphenyl-1,10-phenanthroline (Bphen, 20 nm)/F₁₆CuPc(30 nm)/Al (100 nm)

Deposition rates and thickness of the layers were monitored in situ using oscillating quartz monitors. The evaporating rates were kept at 0.5–1 Å/s for organic layers and 10 Å/s for Al cathode, respectively. Fig. 1 shows the schematic energy level diagram of Device A. The energy level alignments at the ITO and Al electrodes are cited from Refs. [36,37], respectively, while the others from Ref. [38]. The contributions of band bending, interfacial dipole, and chemical reaction to the vacuum level (E_F) shift are not distinguished at the interfaces. Current-voltage (I-V) characteristics of the devices were measured with a Keithley 2400 power supply and were recorded simultaneously with measurements. The forward electric voltage is defined as that the ITO electrode is positive biased. To investigate the interface proprieties of CuPc/F₁₆CuPc, X-ray diffraction (XRD) patterns of the ITO/CuPc/F₁₆CuPc film before and after conductivity transition in Device C with the structure of ITO/CuPc (40 nm)/F₁₆CuPc (40 nm)/poly (ethylene oxide) (PEO)/Al (100 nm) were measured. To carry out these experiments, Al cathode was peeled off. Thin



Fig. 1. Schematic energy level diagram of Device A: $ITO/CuPc/F_{16}CuPc/AI$. All the values shown are in unit of eV. E_{vac} , E_F , LUMO, and HOMO denote the vacuum level, Fermi level, the lowest unoccupied molecular orbital, and the highest occupied molecular orbital, respectively.

PEO layer has been adopted as the cathode buffer layer to improve the performance of organic solar cells [39]. Here it was used as the cathode buffer layer to facilitate the peeling off of the Al cathode by a dilute NaOH solution. Al cathode can be peeled off easily while keeping the CuPc/ F_{16} CuPc interfacial properties unaffected. PEO layer was spin-coated onto F_{16} CuPc with a speed of 2000 rpm from aqueous/isopropanol (1:4 by volume ratio) with a concentration of 3 mg/ml followed by baking in vacuum at 60 °C for 1 h. XRD patterns were measured with a Burker D8 Focus diffractometer using Cu K α radiation ($\lambda = 1.54056$ Å). All the measurements were carried out at room temperature under ambient conditions (temperature of ~20 °C and relative humidity of 20–30%) without encapsulation.

3. Results and discussion

Fig. 2 depicts the J-V curves of Device A: ITO/CuPc/F₁₆-CuPc/Al. The as fabricated device remains in the ON state for the first voltage sweep from 0 to 10 V and for the second one from 0 to -7.6 V. However, with further increase



Fig. 2. *J*–*V* curves of Device A: ITO/CuPc/F₁₆CuPc/Al and Device B: ITO/CuPc/Bphen/F₁₆CuPc/Al.

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