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Nonvolatile memory and opto-electrical characteristics of organic memory devices with zinc oxide nanoparticles embedded in the tris(8-hydroxyquinolinato)aluminum light-emitting layer

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ABSTRACT

The nonvolatile organic memory devices based on the tris(8-hydroxyquinolinato)aluminum (Alq_3) emitting layer embedded with zinc oxide nanoparticles (ZnO-NPs) are reported. The devices have a typical tri-layer structure consisting of the Alq_3/ZnO-NPs/Alq_3 layers interposed between indium tin oxide (ITO) and aluminum (Al) electrodes. An external bias is used to program the ON and OFF states of the device that are separated by a four-orders-of-magnitude difference in conductivity. No significant degradation of the device is observed in either the ON or OFF state after continuous stress ($\sim 10^5$ s) and multicycle ($\sim 10^3$ cycles) testings. These nanoparticles behave as the charge trapping units, which enable the nonvolatile electrical bistability when biased to a sufficiently high voltage. Impedance spectroscopy, capacitance–voltage (*C–V*) and current–voltage (*I–V*) analysis are used to verify the possible physical mechanism of the switching operation. Moreover, it is found that the location of the ZnO-NPs could affect the memory and opto-electrical characteristics of the devices, such as the ON/OFF ratio, threshold voltage and turn-on voltage, which can be attributed to the influence of the ZnO-NPs and diffused Al atoms in the bulk of the Alq_3 layer.

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1. Introduction

Organic materials have attracted considerable attention due to their potential applications in next-generation electronic and optoelectronic devices operating at low powers [1,2]. As one of the significant electronic devices, nonvolatile organic memory device is very promising in next-generation memory applications because of the distinctive advantages of low-power consumption, ultrahigh-density storage, high mechanical flexibility, low cost, and simple fabrication [3,4]. Therefore, with the rapid development of organic memory technology, a lot of effort has been made to improve organic memory device structures and to understand their operating mechanisms.

A number of device structures of the nonvolatile organic memory have been manufactured and extensively explored, such as a single-layer structure consisting of only one type of organic material [5,6], a tri-layer structure in which nano-traps for charge carriers are sandwiched between two organic layers [7,8], and a spin-cast blend of polymer and nano-traps in which nano-traps are randomly dispersed in the polymer layer [9–16]. Because the charges stored inside the nano-traps can efficiently change the conductivity of the organic layer, finding appropriate materials to produce the nano-traps has been the major issue in the tri-layer or hybrid composite structures of the organic memory. So far various kinds of nano-traps materials employed in nonvolatile organic memory devices utilizing sandwich or blend structures have been reported, such as metal nanoparticles [8,10,11], semiconductor nanoparticles [12–14], grapheme [9], fullerene (C_{60}) nanoparticles [15], or core/shell CdSe/ZnS nanoparticles [16].

Among the several types of nano-traps materials, Zinc oxide (ZnO) is a promising material as it is an abundant material with non-toxic, cheap, mechanically flexible and optically transparent [17]. A simple ZnO nanostructure, nanoparticles or nano-powder, has been successfully applied in organic optoelectronics due to their excellent optical, electrical, mechanical and chemical properties [17,18]. For organic memory application, some studies concerning the fabrication and the electrical properties of memory devices with hybrid composites containing ZnO nanoparticles (ZnO-NPs) blended with a polymer layer have been carried out [19,20]. However, studies on the electrical bistabilities, the memory stabilities, and the memory mechanisms in nonvolatile organic memory devices made of ZnO-NPs embedded in a small-molecule layer of tris(8-hydroxyquinolinato) aluminum (Alq_3) layer have not

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been reported yet. The material of Alq_3 has been studied as an organic semiconductor, which are popularly used as an electron transport layer and a light-emitting layer in organic light-emitting diodes (OLEDs) [21]. Organic-light emitting bistable memory devices with a dual function of a nonvolatile memory and light-emitting devices are recently developed for versatile applications [22,23]. It is thus important to investigate on both the electrical switching and opto-electrical characteristics of the Alq_3-based organic memories with the ITO electrode. In this paper, we fabricated and investigated nonvolatile organic memory devices consisting of Alq_3 layers embedded with ZnO-NPs. The ZnO-NPs were deposited within the Alq_3 layers by thermally evaporation. These ZnO-NPs then act as efficient charge storage medium. We characterized the fabricated organic memory devices in terms of their electrical bistability, data retention and cycling endurance properties, and opto-electrical characteristics.

2. Experimental details

The nonvolatile organic memory devices proposed in this study consists of an organic/metal-oxide/organic tri-layer structure interposed between indium tin oxide (ITO) and aluminum (Al) electrodes, as shown in Fig. 1(a). The simple cross-bar memory device was fabricated as follows. First, x-nm-thick Alq_3, 1.5-nm-thick ZnO, and (140-x)-nm-thick Alq_3 thin films were evaporated in sequence onto a cleaned ITO/glass substrate. A very thin ZnO film deposited by thermal evaporation from ZnO powders (purity: 99.99%, purchased from Sigma–Aldrich Co.) is able to form a discontinuous layer of ZnO-NPs on our used organic film. The thickness of the Alq_3 and ZnO layers was controlled by a calibrated quartz crystal oscillator and their deposition rate was maintained at 0.1 and 0.01 nm/s, respectively. Finally, a 100-nm-thick Al thin film was evaporated through a metal mask as the top electrode, resulting in the ITO/Alq_3 (x nm)/ZnO (1.5 nm)/Alq_3 (140 – x nm)/Al (100 nm) structure of organic memory devices. For comparison, the single-layer organic memory devices with ITO/Alq_3 (140 nm)/Al (100 nm) were also fabricated. All materials were evaporated at a vacuum pressure under 3.0×10^{-6} torr at room temperature. The thickness of the films was determined *in situ* by a quartz-crystal sensor and *ex situ* by a surface profiler (Tencor Alpha-step IQ). The active area of the device was 2.5×2.5 mm^2.

The surface morphology and chemical compositions of the films were characterized by atomic force microscopy (AFM, Vecco Instrument, Santa Barbara, CA) and X-ray photoelectron spectroscopy (XPS, VG Scientific, Sigma Probe). The current–voltage (*I–V*) curves, the write-read-erase-read cycles and the data retention characteristics of the devices were measured using a source meter (Keithley 2400) controlled by a computer. Impedance spectroscopy and capacitance–voltage (*C–V*) characteristics were carried out using an impedance analyzer (Wayne Kerr 6500B), whose frequency range was 20 Hz to 20 MHz. Luminance–voltage (*L–V*) characteristics of the devices were measured with a source meter and a luminance meter (LS-100). All measurements were carried out at room temperature under ambient conditions.

3. Results and discussion

3.1. Morphology and compositions of ZnO-NPs

The AFM image for the surface morphology of the 1.5-nm-thick ZnO interlayer deposited on the Alq_3 film is shown in Fig. 1(b). It can be clearly seen that the nanoparticles were randomly distributed on the surface of the Alq_3 layer. In general, layers less than 10-nm-thick show a granular structure and a discontinuous profile. The particle size was approximately estimated in the range of 30–60 nm due to some aggregation of nanoparticles. In addition, the chemical compositions of ZnO-NPs were further examined with XPS and Zn $2p_{3/2}$ spectrum is presented in Fig. 1(c). The Zn $2p_{3/2}$ spectrum peaked at around 1022.0 eV, which can be attributed to the formation of ZnO rather than metallic Zn because the binding energy of the Zn–O bond (1021.9 eV) is higher than that of the Zn–Zn bond (1021.4 eV) [24]. The results confirm that there are ZnO-NPs being created from thermal evaporation.

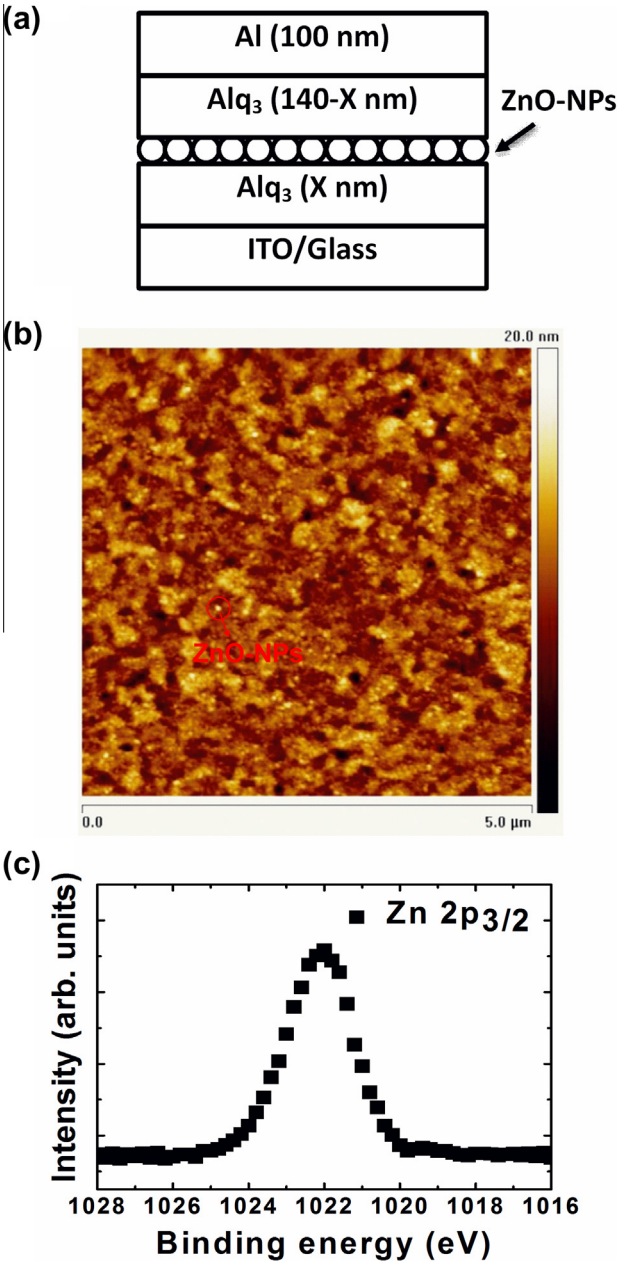

Fig. 1. (a) Schematic structure of the organic memory devices with ZnO-NPs. (b) AFM image and (c) Zn $2p_{3/2}$ cole-level XPS spectra of the 1.5-nm-thick ZnO interlayer deposited on the Alq_3 film.

3.2. I–V characteristics

The typical *I–V* characteristics for the ITO/Alq_3 (70 nm)/ZnO-NPs/Alq_3 (70 nm)/Al (100 nm) device structure were measured from 0 to 10 V and then from 10 to −10 V, as shown in Fig. 2(a). During the first bias scan, a low current was observed in the bias

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