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Optically readout write once read many memory with single active organic layer

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An optically readable write once read many memory (WORM) in Ag/Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH PPV)/ITO is demonstrated in this work. Utilising characteristics of the organic light emitting diode structure of Ag/MEH PPV/ITO and electrochemical metallization of Ag, a WORM with light emitting capability can be realised. The simple fabrication process and multifunction capability of the device can be useful for future wearable optoelectronics and photomemory applications, where fast and parallel readout can be achieved by photons. © 2016 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4940204>]

Resistive switching device has been expected to revolutionize electronic industry through basic concept of the memristor¹ and intrigue memory effect in various kinds of materials ranging from inorganic thin films² to organic films and biology films.³ Recently, resistive switching in organic materials has caught much attention in electronic research due to their intrinsic flexibility, low temperature solution processes, and multifunction capability.⁴ Various types of organic resistive memory materials have shown promising performance for write once read many memory (WORM) and rewritable resistive memory devices.⁵ Although the intrinsic organic materials may play a role in the observed resistive switching related to redox couple, conformation, or coordination chemistry,⁶ majority of cases involve electrochemical metallisation of electrodes.⁷ Through control of the compliance current during biasing, a WORM, rewritable memory, and threshold memory can be realised accordingly in the same device.³ In particular, the WORM memory is an important memory device for a permanent data archive for security purpose where once written, memory state cannot be erased.⁸ Furthermore, the WORM device can be utilized in read only memory in computer for storing boot tasks. It has been witnessed that various innovative device configurations in an organic WORM memory can be realized such as sticker type memory and stretchable memory.⁹

Recent works on resistive memory device are focused on the ability to read memory data through light emission.¹⁰ Reading process in the memory device is serial, where single bit is read after one another, hence it is a slow process. Read out by parallel read process using optical sensor and interconnects can be the solution for fast read out. Tseng *et al.* and Ma *et al.* have shown coexistence of light emitting and memory effect by stacking vertically light emitting layer and memory layer.¹¹ In the same way, Yook *et al.* and Nau *et al.* have demonstrated light emitting memory device using various inorganic layers in series with a light emitting layers.¹² It is important to note that the above complex bilayers light emitting resistive memory can only be achieved with vacuum evaporation of organic layers; while in solution process, organic solvent of the second layer affects the integrity of

the previous layer, hence reports on solution process light emitting resistive memory is scanty. In the WORM device, Wang *et al.* have demonstrated the WORM device with a capability to read optically through random lasing of ZnO layer.¹³ However, optically read-out from a solution-processable single active layer of organic material has not been demonstrated before.

In this work, we demonstrate light emitting capability of the Ag/MEH PPV/ITO device to read memory states. The device shows the WORM behaviour due to electrochemical migration of silver. The simple device structure may pave a way for WORM device on Radio Frequency Identification Device (RFID) with an interactive display and optical read out capability.

Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH PPV) was purchased from Sigma Aldrich and used without further purification. The powder was dissolved in dichlorobenzene and the solution was spin-coated on a clean ITO coated glass. The final film thickness was determined by Atomic force microscopy (AFM) which is around 160 nm. The top Ag electrodes with 0.5 mm² rectangular patterns were evaporated on the film through shadow mask with evaporation rate of 0.005 nm/s. Devices were tested in vacuum at pressure 10⁻⁴ Torr. The topography of the films was studied using a commercial AFM NT MDT model p8. The photoluminescence (PL) spectra were acquired using RF-5301 PC Shimadzu spectrofluorometer. AFM image and PL spectra of the film are shown in the supplementary material (see Ref. 14).

The device configuration is shown in Figure 1(a). In such configuration, ITO electrodes are inert and Ag electrodes are active components.¹⁵ Following the concept of atomic switch, under positive bias applied on Ag electrode, Ag atoms will migrate into the film and connect top and bottom electrode through the metallic filaments formation; therefore, this changes the original device state at low conducting state into high conducting state. When the high compliance current (such as 0.05 A) was chosen, the silver filaments are thick enough such that erasing process by applying negative voltage is not possible to break the filament.

The original device state is in its low conducting state. Only by applying positive voltage, Ag atoms can migrate

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FIG. 1. (a) Schematic picture of the device. (b) OFF state of the device where light emission can take place at -8 V on cathode. (c) ON state after pulsing 8 V where light emission does not take place and electrical current will flow through metallic filaments.

and form a conducting path that gives a high conducting state (written state). With the application of a negative voltage on silver electrode, in the absence of Ag migration, light emission occurs due to the recombination of electrons from silver cathode and holes from ITO anode in this low conducting state. Hence, the low conducting state can be distinguished by light emission; while in high conducting state (written state), there is no light emission because charge is transported through silver filaments instead of organic layer. Figure 1(b) illustrates the mechanism of the optical readable memory. The unique capability of the device is originated from the migration of Ag atoms from top electrodes which play a pivotal role in revolutionizing the resistive memory industry.¹⁵ Furthermore, the work function of Ag is around 4.3 eV which is suitable to function as the cathode for light emission device. Cu is another fast electromigrating metal in

the microelectronic device and memory device; however, its work function is rather high 4.7 eV which cannot be utilised for light emission device.

Figure 2(a) shows the current-voltage (I-V) sweep of the device from 0 V to 8 V (sweep 1). The positive bias of 1.5 V transforms the low conducting state to high conducting state. The current-voltage jump is sharp and abrupt which suggests filamentary mechanism. Reverse voltage sweep from 0 V to -20 V (sweep 3 in Fig. 2(a) only shows up to -10 V) shows a stable high conducting state without any transitions to low conducting state. The I-V shows the WORM characteristic in this silver based organic resistive switching device. We note that there is no electroforming process required in contrast with silver based inorganic resistive switching device.¹⁵ Such contrast can be explained as Ag nanoparticles have been incorporated into organic layers during thermal evaporation and therefore large electroforming is not necessary.⁷

After such transition in Fig. 2(a), the device is switched to ON state permanently and no light emission can be observed at -8 V applied on Ag electrode. The original state of the device (OFF state) is at low conducting state and light emission can be observed at -8 V bias. Long retention of the ON state and OFF state at 0.5 V can be maintained more than 10^4 s and up to 3 weeks. The read endurance can be up to 10^5 reading pulses at 0.3 V with 1 μ s pulse width as shown in Fig. 2(c). Fig. 2(d) shows the intensity of emission light in cd/m^2 as a function of voltage. The photographs of light emission at OFF state are shown in the inset of Fig. 2(d); the orange red emission color is consistent with PL peak in Fig. S2. Light emission is still rather low, due to mismatch of electrodes' work function and organic layer HOMO and LUMO. Although it is possible to enhance light emission by stacking electrons transport layer and hole transport layer,

FIG. 2. (a) WORM memory I-V sweep of the device. (b) Retention of OFF state (original state) and ON state can be retained for longer than 10^4 s. (c) Read endurance of the device. (d) Luminance versus voltage of the device. Inset of (d) shows the optical photograph of light emission at OFF state after applying -8 V on Ag cathode, black square is the Ag electrode area which is opaque to photons.

FIG. 3. (a) Fitting of experimental I-V to charge transport model. (b) Repeatability of different number of devices from different batches. The numbering refers to the device number.

the memory effect could be altered significantly or even suppressed. To understand the charge transport mechanism of the device, we fit the charge transport to theoretical models such as thermionic injection (TI) and space charge limited current (SCLC)

$$I \propto V \quad \text{Ohmic transport,} \quad (1)$$

$$I \propto V^n \quad \text{SCLC transport,} \quad (2)$$

$$I \propto \exp(\sqrt{V}) \quad \text{TI transport.} \quad (3)$$

The high conducting state can be fitted to Ohmic charge transport with $I \propto V$. The Ohmic charge transport implies metallic filaments in the device at high conducting state. The OFF state can be fitted through TI model and SCLC. The fittings are shown in Figure 3. Before threshold voltage at 1.5 V, the charge transport is dominated by the SCLC with $n=4$ in similarity with Ref. 16. The index n is higher than 2 as in our fitting which suggests existence of charge trapping media and also known as trap charge limited current.¹⁷ Despite the absence of nanoparticles as the charge storage medium, as shown in Ref. 16, we can explain the similarity as the charges that are trapped by Ag nanoparticles which were incorporated into the layers during electrode evaporation. At threshold voltage, the nanoparticles connect together and form filaments connecting top electrode and bottom electrode. The low conducting state with the SCLC charge transport is transformed into an Ohmic charge transport in high conducting state. Replication of the effect is shown in Fig. 3(b). Ten devices from 3 different batches show similar I-V and WORM behavior. The band diagram relating to the optically read-out resistive switching is shown in Fig. 4. Initially, the device is in the OFF state (Fig. 4(i)) where electrons and holes injected from

contacts can recombine and emit photons at -8 V read voltage. The diffused Ag atoms created from thermal evaporation form localized energy states in between HOMO and LUMO of MEH PPV. After biasing the device by 8 V, Ag migrations take place and form metal bridge between cathode and anode (Fig. 4(ii)); the device state is switched to the ON state. At -8 V read voltage, there is no light emission when device is in the ON state.

The above described concept may find application in photonics circuits or optoelectronics devices. Furthermore, WORM memory is particularly interesting for data encryption. Here, the typical orange/red light emission organic material, MEH PPV, was used; however, the concept can be expanded to various organic or inorganic materials with narrow emission spectrum such as rare earth ions (Er^{3+} , Yb^{3+}) or security applicable emission wavelength such as THz or

FIG. 4. Band diagram of optically readout resistive switching device.

infrared frequency. Therefore, we envisage that the described concept will enable not only photonics memory but future highly secured data storage as well.

In conclusion, we demonstrate that the WORM memory device with optical read out can be achieved using combination of atomic switching concept and organic light emission. Especially, the simple two terminal device of Ag/MEH PPV/ITO may facilitate applications in the optical memory and optoelectronics. The light emission capability of the device can be further enhanced by utilizing single layer emissive layer such as light emitting electrochemical cell while keeping the memory capability and device simplicity unchanged.

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