Organic bistable light-emitting devices

Liping Ma, Jie Liu, Seungmoon Pyo, and Yang Yang[a)]
Department of Materials Science and Engineering, University of California at Los Angeles, Los Angeles, California 90095

(Received 8 October 2001; accepted for publication 26 November 2001)

An organic bistable device, with a unique trilayer structure consisting of organic/metal/organic sandwiched between two outmost metal electrodes, has been invented. [Y. Yang, L. P. Ma, and J. Liu, U.S. Patent Pending, U.S. 01/17206 (2001)]. When the device is biased with voltages beyond a critical value (for example 3 V), the device suddenly switches from a high-impedance state to a low-impedance state, with a difference in injection current of more than 6 orders of magnitude. When the device is switched to the low-impedance state, it remains in that state even when the power is off. (This is called "nonvolatile" phenomenon in memory devices.) The high-impedance state can be recovered by applying a reverse bias; therefore, this bistable device is ideal for memory applications. In order to increase the data read-out rate of this type of memory device, a regular polymer light-emitting diode has been integrated with the organic bistable device, such that it can be read out optically. These features make the organic bistable light-emitting device a promising candidate for several applications, such as digital memories, opto-electronic books, and recordable papers. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1436274]

Organic electronics has attracted much attention due to the distinctive advantages of organic materials, such as low cost processing,[1] lightweight, mechanical flexibility, etc. Several types of electronic and optoelectronic devices, such as diodes, sensors, and light-emitting diodes, have been made using organic materials for the active medium.[2–4] However, despite impressive progress in organic electronics, there is still a lack of progress in organic electronic memory. Modern digital memory is usually achieved by either electrical, or optical, or magnetic bistable states. The organic electrical bistable device, which usually shows a transition from a high-impedance state to a low-impedance state under an electrical field, have been studied for more than 30 years.[5–9] However, high performance and reliable organic memory devices have not yet emerged, particularly because most organic bistable phenomena to date result from the formation of a conducting filament or an electrical breakdown, which has limited these devices from many applications.

Recently, we reported on an organic electrical bistable device (OBD) having a thin metal layer embedded within the organic material, which is the active medium.[10] The performance of this device makes it attractive for memory cell applications. The two states of the OBD differ in their conductivity by about 10^7 and show remarkable stability in that once the device reaches either state, it remains in that state for a prolonged period of time. More important, the high and low conductivity states of an OBD can be precisely controlled by the application of a positive voltage pulse (to write) or a negative voltage pulse (to erase), respectively. In this letter, we report an organic memory device which combines the electrical bistable characteristics and the electroluminescence of a polymer light-emitting diode (PLED). This combination makes the organic bistable light-emitting device (OBLED) a unique memory device which can be addressed electrically, but the readout is both electrical (in serial) or optical (in parallel). In addition to being a high throughput memory device, this device is also ideal used as an electronic book.

The structures of an OBD and an OBLED are shown in Fig. 1. The organic material used is 2-amino-4,5-imidazoledicarbonitrile (AIDCN). The metal chosen for this illustration is aluminum (Al). Both glass and flexible plastic films were used as substrates to fabricate OBDs. First, the substrates were coated with an 80 nm Al layer. (OBD is a symmetric device, we define the anode to be the electrode on the substrate, and it is wired to the positive potential.) Then, the first AIDCN layer, the middle Al layer, and the second AIDCN layer were sequentially deposited on top of this anode. The device was completed by the deposition of another 80 nm Al layer to serve as the cathode. All depositions were carried out in a vacuum of $\sim 1\times10^{-6}$ Torr in an evaporator equipped with six sources. All the steps, including exchange

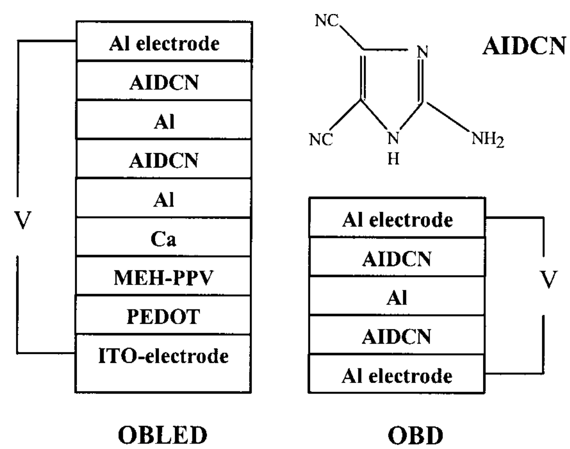

FIG. 1. Structure of an OBD and an OBLED as well as the chemical structure of AIDCN.

[a)]Author to whom all correspondence should be addressed; electronic mail: yangy@ucla.edu

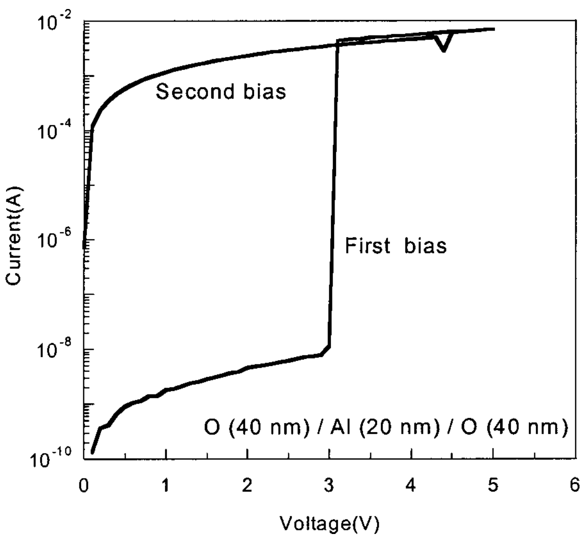

FIG. 2. I–V characteristics of an OBD recorded during the first bias scan and the second bias scan.

of shadow masks for the electrode patterning were conducted without breaking the vacuum. The thickness of both AIDCN layer and the embedded Al layer was about 40 nm and 20 nm, respectively. For the OBLEDs, the devices were constructed by first fabricating a regular PLED with a structure (ITO/PEDOT/MEH-PPV/Ca/Al) in the earlier publication.[11] (MEH-PPV is the abbreviation of poly (2-methoxy-5-(2′-ethyl-hexyloxy)-1,4-phenylene vinylene.) Then, on top of the PLED, an AIDCN/Al/AIDCN trilayer was deposited sequentially. Finally, another Al layer was deposited to finish the device. The forward bias indicates the bottom electrode directly on the substrate is positively biased. Current–voltage (I–V) curves were measured with an Hewlett Packard 4155 semiconductor parameter analyzer. Emission spectra were recorded with a Spectra-scan PR 650.

Typical I–V curves for OBDs are shown in Fig. 2. As can be seen, during the first bias scan, the device as fabricated shows a very low current in the low-voltage range (0–3 Volts), indicating the device is at a high impedance state. However, at a critical voltage, ~3 V in Fig. 2, the current has a sharp increase of nearly 6 orders of magnitude, indicating that the device has had a transition from the high-impedance state to a low-impedance state. When the bias voltage is further increased, the device shows a very high current. However, the I–V curve recorded in the second bias scan is totally different from that observed in the first bias scan. Even in the low-voltage range, the device shows very high current, indicating that the device remains in the low-impedance state. (In fact, the device remains in the low-impendence state even after the power is off; i.e., the non-volatile memory effect.) Thus, these two I–V curves define a voltage range (0–3 Volts in Fig. 2) in which the device shows bistable phenomena, i.e., at a given voltage the device can have two different currents. Most important, the high impedance state can be recovered from the low impedance state by simply applying a reverse bias pulse.[10] This makes OBDs a potential candidate for rewritable digital memories.

One of advantages of using organic materials as the active medium is that devices can be fabricated on flexible substrates, usually plastic films. For example, the success of

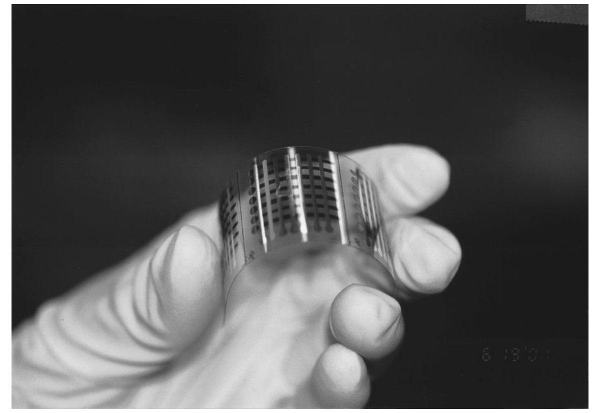

FIG. 3. Photograph of 7×7 organic memory array on a regular plastic overhead transparency.

the light-emitting diodes[12] and transistors[3] fabricated on flexible substrates has been already demonstrated. Figure 3 shows a photograph of three sets of 7×7 OBD matrix fabricated onto a flexible plastic substrate.

Typical I–V curves for an OBLED are shown in Fig. 4, which are similar to the I–V characteristics observed in Fig. 2. During the first sweeping of forward bias, the current has an abrupt increase at a critical voltage, ~6 V. As can be seen in Fig. 4, the current injection "switched up" by 3 orders of magnitude, reflecting a transition of the OBD component from a high-impedance state to a low-impedance state. Meanwhile electroluminescence (EL) can be observed. The inset of Fig. 4 shows the corresponding EL spectrum for an OBLED at a current of 3 mA with the brightness about 280 cd/m^2. However, a different I–V curve as shown in Fig. 4 was observed in the second bias scan. In the low-voltage range(0–6 V), the device has a much higher current injection than that observed in the first bias scan while in the high-voltage range (greater than 6 V), the two I–V curves overlap. Since the current in the second bias scan is limited by the PLED component, the device behaves as a light-emitting diode and EL can be observed at a relative smaller voltage (less than 6 V).

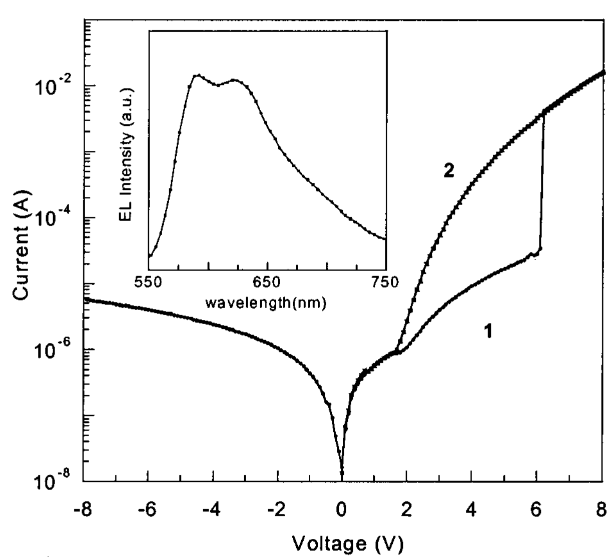

FIG. 4. I–V characteristics of an OBLED (curve 1 is for the first bias scan and curve 2 is for the second bias scan). The inset is the EL spectrum.

The two important features observed in the OBLED—bistability and EL make the OBLED a promising candidate for memories with a high output rate. With OBLEDs, the memory effect can be achieved by first applying a recording voltage pulse (e.g., 8 V in Fig. 4) greater than the critical voltage to trigger the transition from a high-impedance state to a low-impedance state. A small reading voltage (e.g., 4 V in Fig. 4), smaller than the critical voltage, can be applied to OBLED, and the detection of the memory state can be done either electrically (measuring the injection current) or optically (measuring light emission). For optical detection, the devices/pixels at low impedance can give much stronger light emission than those at high impedance because of the difference in current injection. Thus, the recorded or memorized information can be extracted by distinguishing the intensity of light emission at different pixels.

For traditional OBD memory devices having the X–Y matrix format, the reading of the memory array is usually in serial sequence, i.e., the maximum data throughput is achieved by scanning one line after another. However, the optical detection capability of OBLEDs enables parallel reading, which will enhance the reading speed dramatically; the throughput of the memory array can become instant by turning on the whole memory array simultaneously. Most important, due to the presence of the EL, human eyes can be used as to detect (read) the recorded information directly, which further offers OBLEDs new potential applications, such as electronic books, newspapers, smart papers, etc.

In summary, it has been demonstrated that OBDs can be made by sandwiching an organic/metal/organic trilayer between two electrodes. The as-fabricated devices show a sharp increase (6 orders of magnitude) in current injections when the bias voltage reaches a critical value, indicating that the devices have a transition from a high-impedance state to a low-impedance state. Furthermore, a type of memory device, i.e., organic bistable light-emitting devices, which show both electrical bistability and EL, can be made by integrating the bistable device with a regular organic light-emitting diode. For these devices, the memory effects (or recording-reading capability) are realized by first applying a large voltage pulse to record information electrically, and then applying another small voltage pulse to read it either electrically (measuring currents) or optically (measuring the EL).

[1] D. Braun and A. J. Heeger, Appl. Phys. Lett. **58**, 1982 (1991); J. Bharathan and Y. Yang, *ibid.* **72**, 2660 (1998); D. A. Pardo, G. E. Jabbour, and N. Peyghambarian, Adv. Mater. **12**, 1249 (2000); T. F. Guo, S. C. Chang, S. M. Pyo, G. F. He, and Y. Yang (unpublished).
[2] C. W. Tang and S. A. Van Slyke, Appl. Phys. Lett. **51**, 913 (1987).
[3] F. Garnier, R. Hajlaoui, A. Yassar, and P. Shirakawa, Science **265**, 1684 (1994).
[4] F. Hide, M. A. Diaz-Garcia, B. J. Schwartz, M. R. A. Andersson, Q. Pei, and A. J. Heeger, Science **273**, 1833 (1997).
[5] J. Kevorkian, M. M. Labes, D. C. Larson, and D. C. Wu, Discuss. Faraday Soc. **N51**, 139 (1971).
[6] R. S. Potember, T. O. Poehler, and D. O. Cowan, Appl. Phys. Lett. **34**, 405 (1979).
[7] R. Kumai, Y. Okimoto, and Y. Tokura, Science **284**, 1645 (1999).
[8] L. P. Ma, W. J. Yang, Z. Q. Xue, and S. J. Pang, Appl. Phys. Lett. **73**, 850 (1998).
[9] J. Chen, M. A. Reed, A. M. Rawlett, and J. M. Tour, Science **286**, 1550 (1999).
[10] L. P. Ma, J. Liu, and Y. Yang, U.S. Patent Pending, U.S. 01/17206 (2001); *News of the Week*, Science, September 7 (2001), p. 1746.
[11] J. Liu, Y. Shi, and Y. Yang, J. Appl. Phys. **88**, 605 (2000).
[12] G. Gustafsson, Y. Cao, G. M. Treacy, F. Klavetter, N. Colaneri, and A. J. Heeger, Nature (London) **357**, 477 (1992).