



Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

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Version of record first published: 18 Oct 2010

To cite this article: Liping Ma, Jie Liu, Seungmoon Pyo, Qianfei Xu & Yang Yang (2002): Organic Bistable Devices, *Molecular Crystals and Liquid Crystals*, 378:1, 185-192

To link to this article: <http://dx.doi.org/10.1080/713738589>

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Organic Bistable Devices

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A novel organic bistable device with an unconventional structure, i.e., an organic/metal/organic triple layer sandwiched between two outmost metal electrodes, has been developed. The current-voltage characteristics have a sharp increase in injection current by as much as 6 orders of magnitude when the applied bias is larger than a critical voltage. After having had the electrical bias, the device remains in the low impedance state even when the power is off. (This is called the "nonvolatile" phenomenon in memory devices.) The high impedance state can be recovered by applying a reverse bias; therefore, this controllable electrical bistability is ideal for memory applications. Similar electrical bistable behavior has been observed in devices using several different metals and organic materials. Our results suggest that the unique device structure, i.e., a thin metal layer embedded between two organic layers, is critical for the observed bistability. Discussions of the possible operational mechanisms are provided.

Keywords: organic electronics, organic memory, organic devices

Several types of organic electronic and opto-electronic devices have been developed using organic functional materials as the active compound. For example, light-emitting diodes (LEDs), thin film transistors, solar cells, etc. have been reported [1,2,3]. However, one important electronic device—high performance organic memory—is still missing from the list of organic electronic devices, although switching and memory phenomena in organic materials and devices have been reported for more than 30 years [4,5,6,7,8]. Previous observations of organic bistable phenomena were mainly due to association with the formations of conducting filaments, which have been found not to be reliable for long-term operation.

Recently, it has been found that when a metallic thin film is embedded between two organic layers, the resultant device shows a unique electrical bistability [9]. The two different states of the electrical bistability differ in their conductivity by as much as 10^6 and show a remarkable retention,

Received 10 October 2001; accepted 8 November 2001.

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i.e., once the device reaches either state, it tends to remain in that state for a prolonged period of time. In this manuscript, we further explore this unique electrical bistability by fabricating organic bistable devices (OBDs) with different layers of embedded metals and organic materials. In addition, the temperature and area dependence of the injection current of OBDs are also presented. The initial results strongly suggest that the operating mechanisms of OBDs are likely not due to filament formation.

The OBD shown in Figure 1 consists of an organic/metal/organic triple layer sandwiched between two outermost metal electrodes. This manuscript reports experimental results observed for devices fabricated from different organic compounds and different metals. The organic compounds we have tried are 2-amino-4,5-imidazoledicarbonitrile (AIDCN, inset of Figure 2a), tris-8-(hydroxyquinoline) aluminum (Alq3, inset of Figure 3a) and poly(2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene (MEH-PPV, inset of Figure 3b). For the middle metal layers, we have tried aluminum, copper, silver, and gold. The devices were all fabricated by thermal evaporation under a vacuum of $\sim 1 \times 10^{-6}$ torr without breaking the vacuum. The detailed fabrication was described elsewhere [10]. In brief, the fabrication starts with the deposition of the Al electrode (80 nm) on a piece of pre-cleaned substrate, subsequently followed by the sequential deposition of the first organic layer, the embedded middle metal layer, and the second organic layer. Finally, another Al layer (80 nm) was deposited as the cathode. The area of the device, which is defined as the overlap of the anode and the cathode, is 1 mm^2 unless further specified. The organic layers and the embedded metal layer all have a thickness in the nanometer range. Current-voltage (I-V) curves were measured with a Hewlett Packard 4155 semiconductor parameter analyzer in ambient environment. A low temperature dewar VPF-475 equipped with a temperature controller was used in measuring the I-V characteristics of OBDs at different temperatures.

Typical I-V curves of an OBD, for example for a AIDCN(40 nm)/Al(20 nm)/AIDCN(40 nm) device, are shown in Figure 2. During the first voltage scan in the low voltage region (0–3 volts), very low current injection was observed. However, at a critical voltage, the current sharply increased

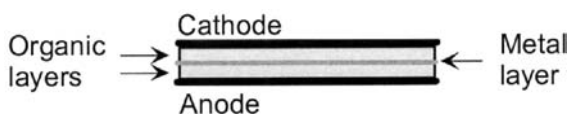


FIGURE 1 The device structure of a typical organic bistable device (OBD).

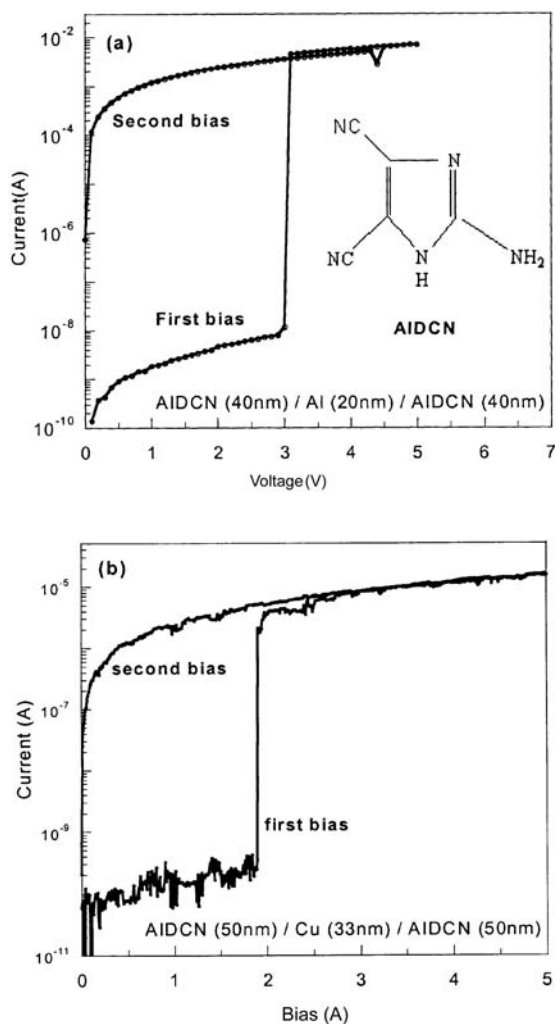


FIGURE 2 Current-voltage characteristics of OBDs fabricated with AIDCN and (a) Al, (b) Cu, (c) Au, and (d) LiF, respectively.

by 6 orders of magnitude, from 10^{-9} amperes to about 10^{-3} amperes, indicating the device had a transition from a high impedance state ("OFF state") to a low impedance state ("ON state"). Above the critical voltage, the current increases nearly linearly with increasing bias voltage. However, the second voltage bias scan yielded a different I-V curve. In the low voltage region, the current injection was several orders of magnitude greater than that observed during the first bias scan, thus confirming that the device

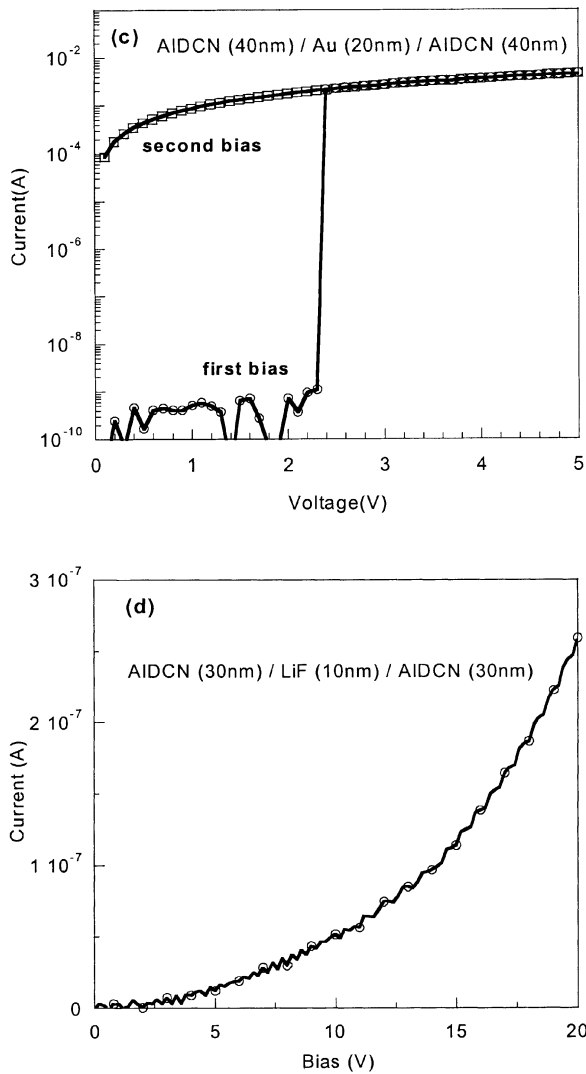


FIGURE 2 (Continued)

remained in the low impedance state even though the power was off. This retention of low impedance (as well as the high impedance) state is ideal for nonvolatile memory applications. After this transition, the device remains in this state even after the power is turned off, as can be seen in the second voltage scan. The two I-V curves in Figure 2a define the electrical bistability of the OBD (between 0 and 3 V) and also reveal the nonvolatile nature of the

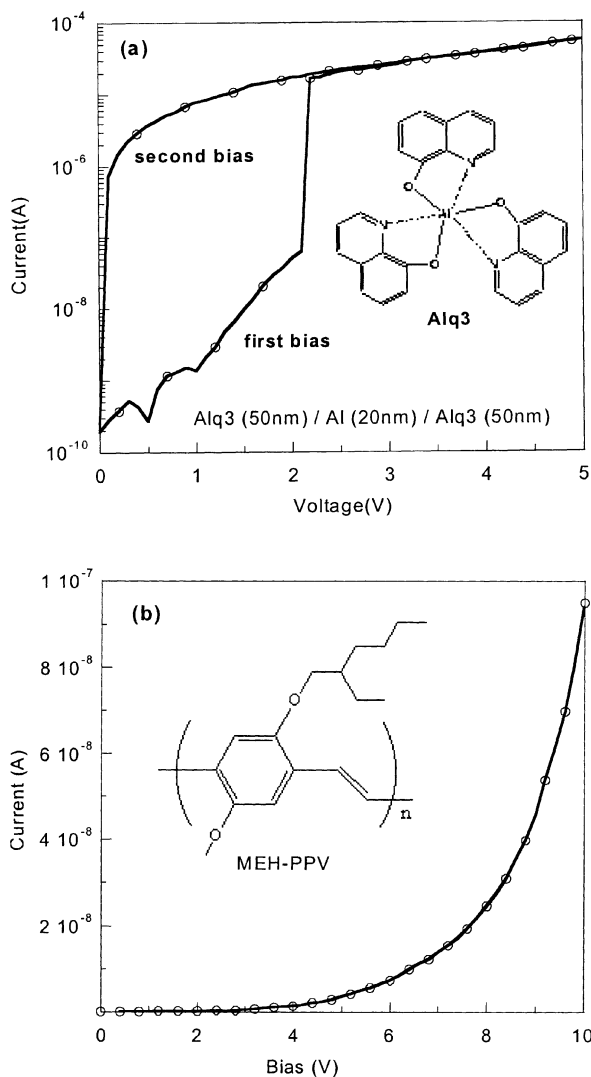


FIGURE 3 Current-voltage characteristics of OBDs fabricated with Al and different organic materials (a) Alq3 and (b) MEH-PPV, respectively.

memory. One of the most important features of our OBD is that the OFF state can be recovered by the simple application of a reverse voltage pulse [9,10]. This is equivalent to the “erasing” process of a digital memory cell. However, the bistable behavior, the OFF-ON transitions, and the creation of nonvolatile memory effects can be observed only in the presence of the

embedded thin metal layer. The switching time for the OBD is in the nanoseconds time scale.

It is well known that in organic light-emitting diodes the active metal electrodes, such as Al, will often chemically react with organic compounds and form a metal-organic complex at the interface. It is not clear whether and how (if any) the formation of those complexes contribute to the observed electrical bistability. However, similar switching and memory behavior has also been observed when the embedded Al layer is replaced by less reactive metals, such as Cu (Figure 2b), Ag, and especially Au, which is fairly inert (Figure 2c). In addition, we purposely replaced the middle metal layer with a thin layer of lithium fluoride (LiF), an electrically insulating material, and there was no electrical bistability observed (Figure 2d).

The criteria for materials selection are still not well established. Initial results suggest that organic materials with high dielectric constants and low electrical conductivity show relatively better performance, at least in the thickness range we studied. In addition to AIDCN, several different organic materials and polymers were used to fabricate OBDs successfully. For example, devices using Alq3, which has been widely used to fabricate organic light-emitting diodes (OLEDs) [11], as organic layers instead of AIDCN also show similar electrical bistable phenomena, as can be seen in Figure 3a. The conductivity of the organic compound seems to play an important role in determining the electrical and memory effects. The bistable behavior can only be observed when the organic compound has quite low electrical conductivity. For example, when an AIDCN layer was replaced by either MEH-PPV (a polymer frequently used to fabricate polymer light-emitting diodes) (Figure 3b) or C₆₀ (a conductive organic material), no electrical bistable phenomena could be observed.

Failure to observe bistability in the AIDCN and Alq3 single-layer (anode/organic/cathode) devices and the AIDCN/LiF/AIDCN device suggests that the switching and memory phenomena are closely associated to the distinct structure of OBDs, i.e., the presence of the metal layer embedded between two organic layers. The detailed mechanisms involved in the OBDs are still under investigation. However, initial results have provided strong evidence that the mechanisms of our OBDs are most likely not due to the formation of conducting filaments [4,5], which have made most organic-bistable devices reported to date very controversial. If metal filaments were involved, metallic behavior—the metallic I-V characteristics of OBDs in the ON state—would be expected and the magnitude of the current would increase as the temperature decreased [4,12]. However, as shown in Figure 4, the current was found to decrease as the temperature decreased.

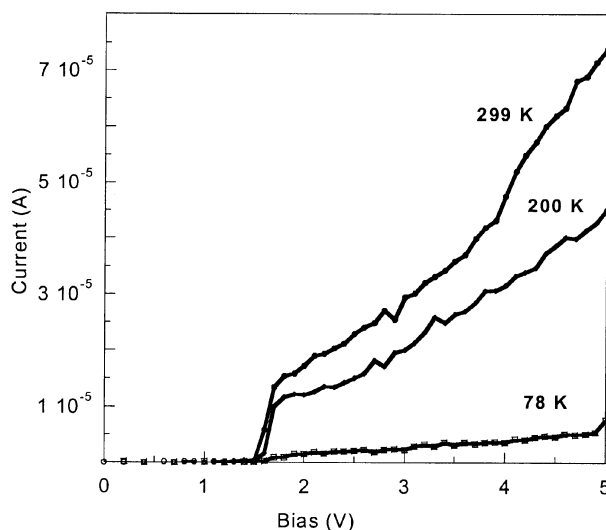


FIGURE 4 Current-voltage characteristics of an OBD measured at 299 K, 200 K, and 78 K.

In addition, if the formation of conducting filaments had been involved, the injected current would be insensitive to the device area [4,5]. Or, if they had been involved the currents should show a random dependence, because the dimensions of the filaments reported earlier have been found to be much smaller compared to the devices' area. However, for OBDs reported here, the magnitude of current injection observed in the ON state was nearly proportional to the device's area.

In summary, a new type of device, an organic bistable device, has been developed, and it has a unique structure, i.e., an organic/metal/organic triple layer sandwiched between two metal electrodes. The existence of two different impedance states, shown as two different current injections by up to 6 orders of magnitude under the same voltage bias, makes this type of device an ideal candidate for memory applications. The use of OBDs as digital memory devices also has several advantages. First the bistability, as shown in Figure 2, can be detected at voltages of less than 1 volt since the current varies from the microampere range for the low impedance state to the pico/nanoampere range in the high impedance state, thus requiring a very low power consumption estimated to be in the submicrowatt range. Second, unlike their inorganic counterparts, which utilize expensive systems such as chemical vapor deposition to grow the thin films, OBDs can be prepared using relatively low cost equipment and processes, such as thermal

evaporation. In addition, since they have the advantages of using organic materials, there is the potential to fabricate large area, light-weight, and flexible memory devices.

ACKNOWLEDGEMENTS

This research is partially supported by the Air Force Office of Scientific Research (Grant # F49620-00-0103), and the Office of Naval Research (Grant # N00014-01-1-0136). We would like to dedicate this invention to Prof. Sukant K. Tripathy of University of Massachusetts-Lowell for his outstanding contribution in the field of functional polymers. Prof. Tripathy passed away in an accident on December 12, 2000.

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