Electrical characteristics of an organic bistable device using an Al/Alq3/nanostructured MoO3/Alq3/p+-Si structure

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The electrical properties of a device with an Al/Alq3/nanostructured MoO3/Alq3/p+-Si structure were investigated for organic resistance switching memories. The conductance of the device can be electrically switched to either high conductance or low conductance. The bistable switching of the device is attributed to the MoO3 nanocluster layer interposed between the Alq3 thin films. When the device was switched to high conductance, a space-charge field dominated carrier transportation of the device. The space-charge field was resulted from charges trapped in the MoO3 nanoclusterlike layer. Both retention measurement and write-read-erase-read cycles of the device are also provided. © 2010 American Institute of Physics. [doi:10.1063/1.3299265]

Interest in organic memories1–21 has rapidly increased due to promise of better scalability, low-cost fabrication, mass production capability, and mechanical flexibility. Many published results have focused on organic resistance switching memories with nanostructured materials as charge trapping centers.2,4,7–12,14,15,17,19–21 Because of the distinct properties of the nanostructured materials, organic resistance switching memories with high density, large ON/OFF ratios, and other superior performances will be obtained in the near future. The nanostructured materials can be composed of a nanocluster/organic-metal composite layer fabricated by a thermal evaporation method2,4,8 (e.g., aluminum cores covered with aluminum oxide shells)3. The nanostructured materials can also be composed of a nanocomposite of polymer-gold nanoparticles fabricated by chemical synthesis.7,9,10 (e.g., 2-naphthalenthiol capped gold nanoparticles). In addition, metal nanoparticles,8,15 oxide nanoparticles,11,19–21 C60 nanoparticles,12,14 or core/shell type CdSe/ZnS nanoparticles17 have been introduced to be the nanostructured materials.

Molybdenum trioxide (MoO3) has been extensively applied in organic optoelectronics (e.g., as a doping layer to raise conductivity,22 a buffer layer to increase carrier injection23 or carrier collection,24 an intermediate layer to protect underlayers,25 or a capping layer to enhance light coupling24). Such wide applications are attributed to the characteristics of the MoO3 thin film: high work function, high conductivity for holes, and high transparency. In this study, we used a nanostructured MoO3 layer as the charge storage layer of an organic bistable device (OBD). The OBD consists of an Al/Alq3/MoO3/Alq3/p+-Si structure. The MoO3 layer not only is a good hole trapping center, but also can be deposited easily at low temperature by thermal evaporation. The OBD exhibited a high ON/OFF current ratio up to 105, long retention time over 4000 s, and a rewritable/erasable feature. The formation of the bistable switching of the OBD is ascribed to the charge trapping effect of a MoO3 nanoclusterlike layer. Moreover, the current-voltage (I-V) characteristics of the OBD were quite different from those of an OBD using a MoO3 nanoparticles layer.20 No negative differential resistance (NDR) was observed in the I-V characteristics of the OBD. This phenomenon probably results from the dissimilar surface morphology of the MoO3 layer deposited on the Alq3 thin film. Because of both the simple structure of the OBD and the merits of the Alq3 (e.g., low cost, easiness of preparation, and high stability), complexity of fabrication and the production cost are greatly reduced. Furthermore, the OBD can be easily embedded into the well-developed semiconductor fabrication processes.

The OBD proposed in this study consists of an organic/metal-oxide/organic tri-layer structure interposed between top and bottom electrodes. First, 50 nm thick Alq3, 5 nm thick MoO3, and 50 nm thick Alq3 thin films were evaporated in sequence onto a cleaned p+-type silicon substrate. The average deposition rate of the Alq3 thin films and that of the MoO3 layer were about 0.1 and 0.01 nm/s, respectively. Finally, a 100 nm thick Al thin film was evaporated through a metal mask with 2 mm×2 mm square patterns as the top electrode. All materials were evaporated at a vacuum bellow 3×10^-6 torr at room temperature. The I-V characteristics of the OBD were measured using a Hewlett Packard 4156A semiconductor parameter analyzer in ambient environment. The Al electrode was kept at 0 V, and all bias conditions were applied on the p+-Si substrate. The composition of the Alq3/Al interface was analyzed using an x-ray photoemission spectroscopy (XPS). The surface morphology of the MoO3 layer on the Alq3 thin film was analyzed using an atomic force microscope (DI-Veeco Instrument).

The I-V characteristics of an OBD with an Al/Alq3/nanostructured MoO3/Alq3/p+-Si structure are shown in Fig. 1(a). At first sweeping bias (the olive curve), a sweeping bias from 0 to 10 V was applied with 100 mA current compliance. Initially, the OBD was in a low conductance state which had a current level of 10^-10–10^-5 A. An abrupt increase in the current level was observed when the applied voltage was about 3.5 V. Then, the OBD held at a high conductance state when the applied voltage swept to higher voltage. It is clear that the OBD underwent an elec-
The bistability of an OBD with an Alq3/p+-Si structure is attributed to the Al–O compound near an Al electrode. To verify this, we used Ag to replace Al as the top electrode. The bistability of the OBD was again recognized from the I-V characteristics of an OBD composed of a 100 nm Ag/100 nm Alq3/p+-Si structure.

FIG. 2. (Color online) Al (2p) XPS curve of the Al/Alq3 interface of the OBD.

The bistable switching of the OBD is referred to the nanostructured MoO3 layer sandwiched between the Alq3 thin films. But some published results suggested that an Al/O compound was generated at the interface between Al and Alq3 thin films. Namely, the bistable switching of the OBD, the trapped holes in the charge trapping centers were released. The OBD was restored to the low conductance state again when a following sweeping bias from 0 to 10 V served as “writing” and “erasing” processes, respectively. Thus, the OBD holds the essences of a memory: bistability and rewritability.

The bistable switching of the OBD is referred to the nanostructured MoO3 layer sandwiched between the Alq3 thin films. But some published results suggested that an Al–O compound near an Al electrode(s) is responsible for the conductance switching of OBDs with an Al electrode(s). As can be seen in Fig. 2, an Al–O compound was generated at the interface between Al and Alq3 thin films. Namely, the bistable switching of the OBD could be relative to the Al–O compound. To verify this, we used Ag to replace Al as the top electrode. The bistability was again recognized from the I-V characteristics of an OBD with an Ag/Alq3/nanostructured MoO3/Alq3/p+-Si structure, as shown in Fig. 1(b). For comparison, the I-V characteristics of an OBD composed of a 100 nm Ag/100 nm Alq3/p+-Si structure are shown in the inset of Fig. 1(b). No hysteresis as obtained in the I-V characteristics of the OBDs with nanostructured MoO3 layer was observed. Therefore, we can conclude that the nanostructured MoO3 layer dominates the bistability of an OBD with an Al/Alq3/nanostructured MoO3/Alq3/p+-Si structure.

The ON/OFF state switching of the OBD, with an Al/Alq3/nanostructured MoO3/Alq3/p+-Si structure, is resulted from hole trapping/de-trapping provided by the nanostructured MoO3 layer. At the low applied voltage of the first sweeping bias in Fig. 1(a), the current was very small. Most of holes injected from the p+-Si into the OBD were trapped by charge trapping centers given by the nanostructured MoO3 layer. As a result, the OBD stayed at the low conductance state. By applying a voltage above the threshold, numerous holes were injected into the OBD, and the charge trapping centers were filled sufficiently. Then, the nanostructured MoO3 layer was polarized, and the interfacial resistance of the nanostructured MoO3 layer was decreased. The OBD was switched to the high conductance state. On the contrary, while an adequate reversed bias was applied on the OBD, the trapped holes in the charge trapping centers were released. The OBD was restored to the low conductance state.

By fitting the I-V curve in a log-log scale of the high conductance state of the OBD, linear relation between current and voltage was found, as shown in the inset of Fig. 1(a). Such relation indicates that carrier transportation is influenced by space charges at the high conductance state. In addition, NDR observed in the I-V characteristics of OBDs using a MoO3 nanoparticles layer was not obtained in Fig. 1(a). Such dissimilarity probably originated from the surface morphology of the MoO3 layer. As shown in Fig. 3, the surface morphology of a 5 nm thick MoO3 layer grown on the Alq3/p+-Si was not a nanoparticle feature, but a nanoclusterlike feature. Polarization resulted from charge trapping of the MoO3 nanoclusterlike layer was more random than that of the MoO3 nanoparticles layer. This means the effective polarization effect on the carrier transportation of the MoO3 nanoclusterlike layer is weaker than that of the MoO3 nanoparticles layer. Therefore, after switched from the low conductance state to the high conductance state, the OBD maintained at the high conductance state without NDR. These results also suggest that the surface morphology of a nanostructured MoO3 layer plays an important role in the electrical characteristics of OBDs using nanostructured MoO3.
Two important properties of the OBD are shown in Fig. 4, retention time and rewritable/re-erasable ability. The retention measurement of the OBD was carried out by applying a bias at 1 V, as shown in Fig. 4(a). There was no appreciable change in the current of the high conductance state. Oppositely, current fluctuation occurred in the low conductance state. The current fluctuation of the low conductance state is because of the incomplete erasing of trapped charges after an erasing process. Nevertheless, the current of the high conductance state was getting smaller and more stable with time, and the ON/OFF current ratio became larger. Overall, a clear conductance difference between ON/OFF states could be recognized, and our device possessed long retention time after an erasing process. Nevertheless, the current of the low conductance state was getting bigger and more fluctuant in the low conductance state.

In summary, the electrical properties of an OBD with an Alq3/nanostructured MoO3/Alq3/p+-Si structure were studied. We showed that the bistability of the OBD is derived from the charge trapping effect of the MoO3 nanoclusterlike layer interposed between the Alq3 thin films. Conductance switching without NDR was observed in the I-V characteristics of the OBD, which differs from that of an OBD with a MoO3 nanoparticle layer. The electrical characteristics of OBDs using nanostructured MoO3 are largely affected by the surface morphology of a nanostructured MoO3 layer.

FIG. 3. (Color online) Surface morphology of the 5 nm thick MoO3 layer deposited on the Alq3/p+-Si structure were.

FIG. 4. (Color online) (a) Retention measurement of the OBD. The pink (upper) and green (lower) circles correspond high and low conductance states, respectively. (b) The reading currents after writing and erasing of the OBD for the first eight cycles. The red (upper) and olive (lower) circles correspond high and low conductance states.