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Nonvolatile memory effect of an Al/2-Amino-4,5-dicyanoimidazole/Al structure

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Abstract

We studied electrical properties of 2-Amino-4,5-dicyanoimidazole (AIDCN), which has a very large electric dipole moment, using a simple Al/AIDCN/Al structure. The measured current-voltage characteristics showed that the device is switched from low-conductivity state (off-state) to high-conductivity state (on-state) at threshold voltage of 2.3 V. On/off ratio of the current was 10³ at the bias of 1V. The device remained the on-state for two weeks even when the bias voltage was turned off. Furthermore, the on-state could be switched back to the off-state by applying a negative bias voltage. The switching time for transitions was estimated to be below 40 ns by using the transient technique. These results demonstrate that a nonvolatile memory device can be realized by the simple MIM structure. We will also show the temperature dependence of the electrical conductivity, ultraviolet photoemission spectra (UPS) to discuss mechanisms of the switching phenomena.

Keywords: Transport measurements, Organic/inorganic interfaces, Photoelectron spectroscopy

Introduction

There has been increasing interest in developing organic-based electronic devices as organic light emitting diodes, organic field effect transistors, and organic switching devices. Since the organic devices involve metal electrodes for carrier injection, studies of the interface between metal and organic thin films are of crucial importance in revealing electrical properties. Ultraviolet photoemission spectroscopy (UPS) have been a powerful method in investigating the interfacial electronic structure and the energy level alignment between metals and organic thin films[1-7]. 2-Amino-4,5-dicyanoimidazole (AIDCN) has a very large electric dipole moment of 9.13 debye[8] derived from electron donating nature of amino group and electron accepting nature of cyano group. The chemical structure of AIDCN molecule is given in Fig.1.

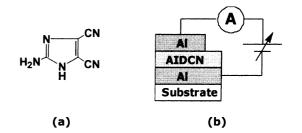


Fig.1. (a) The chemical structure of AIDCN. (b) The structure of nonvolatile memory device.

Recently, Ma et al. reported a nonvolatile memory device consisting of three layers, AIDCN/Al/AIDCN, between top and bottom Al electrodes (Al/AIDCN/Al/AIDCN/Al)[9]. The device has two conductivity states that can be set at two different voltages, and read by measuring the current at a lower voltage.

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Recently, Kawakami *et al.* reported a switching phenomenon in an Al/AIDCN/Al structure[10]. However, a nonvolatile effect has not been observed in the structure. In the present study, we report the *I-V* characteristics of an Al/AIDCN/Al structure and show the nonvolatile memory effect is realized by the simple MIM structure. We further measured UPS spectra of an AIDCN film on oxidized Al substrate and an Al-deposited AIDCN film to discuss the energy level of AIDCN film sandwiched by Al electrodes.

1. Experimental

Commercially obtained AIDCN (Tokyo Kasei) was used without further purification. A 50-nm-thick Al electrode was evaporated on a clean glass substrate through a metal mask at a rate of 0.3 nm/s. After exchanging of the metal mask in the air, 60-nm-thick AIDCN layer was deposited onto the Al layer at a rate below 0.02 nm/s. The Al/AIDCN/Al structure was then obtained by vacuum deposition of a top electrode of 50-nm-thick Al through another metal mask onto the AIDCN film at a rate below 0.02 nm/s. The film thickness of each layer was estimated with a quartz thickness monitor and a Dektak profilometer. The AIDCN and Al films were deposited on the substrate at room temperature and a base pressure of 2×10^{-6} and 8 $\times 10^{-6}$ Torr, respectively. The *I-V* measurement was carried out using a Keithley 2400 sourcemeter in 1-atm nitrogen environment at room temperature. The sweep speed of the applied voltage was 0.025-0.1 V/sec. Preventing the device from break down by excessive current, the current limiter was set at 5×10^{-6} A in a positive bias region. The dc electrical conductivity of the device was also measured with a Keithley 487 picoammeter and an Advantest R6551 power supply, and recorded in a temperature range of 80-295 K using a two-probe technique. The sample was placed in a liquid nitrogen cryostat in which temperature was monitored through a calibrated Si diode sensor located in close proximity to the sample. The conductivity σ at each temperature was determined by an ohmic region in I-V curves. He I UPS spectra of AIDCN on oxidized Al film (AIDCN/AI) and Al-deposited AIDCN films (Al/AIDCN) were measured by using an ultrahigh vacuum (UHV) photoelectron spectrometer with a 180° hemispherical analyzer[11]. Spectra were recorded with -4 V bias on the sample to clear the vacuum level of the sample. Al and AIDCN were evaporated in the preparation chamber of base pressure ~10⁻⁹ Torr. For AIDCN/Al, 10-nm-thick Al was deposited on a Cu substrate, and then oxidized by exposing the evaporated Al film to the air for 10 minutes to obtain Al₂O₃ layer that exists in the device.

AIDCN was evaporated on the Al_2O_3 layer. Al/AIDCN samples were prepared on a clean highly oriented pyrolytic graphite (HOPG). The thickness of each layer was monitored by a quartz thickness monitor. The position of the Fermi level (E_F) was determined from UPS of evaporated Au films.

2. Results and Discussion

The I-V characteristics of the device are presented in Fig. 2. When the applied bias is in the range of 0-0.5 V, the current is very low and follows ohm's low. At the higher voltage, the current increases gradually as receding from ohm's low, and jumps suddenly at 2.3 V by 3 orders of magnitude (scan (1) in Fig. 2). Once switched to the high-conductivity state, the current under the reverse scan (2) (from 3.5 to 0 V) is higher than that of the forward scan (1). We found that the device remained high-conductivity state for two weeks even when the bias voltage was turned off. In the negative bias scan (3), the conductivity state was switched to low-conductivity state at -1.3 V. These results were reproducibly observed, indicating that the device is switched from low conductivity state to high-conductivity state by carrier injection, demonstrating that a nonvolatile memory device can be realized by the simple MIM

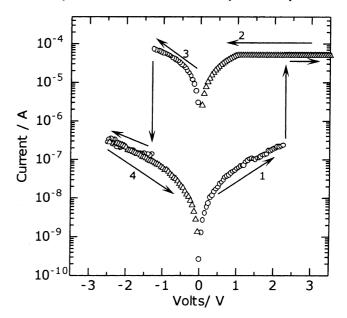


Fig.2. The *I-V* characteristics of Al/AIDCN/Al. The arrows and numbers guide the direction of current variation with voltage. The open circle and the triangle show upward and downward scan, respectively.

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