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Operation Mechanism and Novel Functions of Oxide-Based Atomic Switches

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Abstract

We have investigated the operation mechanism and resistive switching characteristics of Cu(Ag)/Ta_2O_5/Pt atomic switch-type resistance change memories. Ionic current associated with redox reactions at Cu(Ag)/Ta_2O_5 interfaces was clearly observed, and it was found that moisture absorption in the oxide matrix contributes significantly to redox processes as well as switching behavior. In addition to bi-stable switching, novel functions such as conductance quantization and synaptic plasticity were demonstrated, which can be used for neuromorphic applications.

1. Introduction

Among several emerging technologies for the next-generation nonvolatile memory, resistive switching memory based on metal ion transport in a thin oxide layer is one of the most attractive candidates, because of its promising properties such as simple structure, excellent scalability, ease of operation, and high compatibility with the current CMOS fabrication processes [1]. The basic structure of the devices consists of metal-ion conductor-metal (MIM) cells, in which an ion transport layer is sandwiched between an electrochemically active metal electrode (usually, Cu or Ag) and an inert metal electrode (for example, Pt). Because its operation mechanism is essentially the identical to that of an 'gap-type atomic switch', whose resistance across a nanometer gap between a mixed conductor electrode and an inert electrode is controlled by the formation and annihilation of a metal bridge under electrical bias [2], the MIM-structured cell with an active electrode can be referred to as a 'gapless-type atomic switch' [3]. This type of cells is also called an 'electrochemical metallization (ECM) cell' and a 'conductive-bridge random access memory (CBRAM).

Here, we present our recent results on the operation mechanism of atomic switches with a Ta_2O_5 layer as a model system and their novel functions.

2. Redox reactions at metal/oxide interfaces

Ta_2O_5-based atomic switch cells are fabricated on a SiO_2/Si substrate. First, Ti and Pt are deposited by electron-beam (EB) evaporation as the adhesion and bottom electrode, respectively. Then, a Ta_2O_5 layer with thicknesses of 8 – 15 nm is deposited by RF sputtering or EB evaporation. Finally, Cu or Ag is deposited as the top electrode covered with a protective layer of Pt. The cell consists of a cross-point structure with junction sizes between 50 μm and 500 nm.

Cu(Ag)/Ta_2O_5/Pt cells show bipolar resistive switching under bias voltage sweeping. They are SET from a high-resistance (OFF) state to a low-resistance (ON) state at positive bias to the Cu(Ag) electrode, and RESET from the ON state to the OFF state at negative bias. From the measurements of the forming time and the thermal stability of ON states, we proposed the switching mechanism [4]. The SET process corresponds to the formation of a metal filament by nucleation and growth of the active metal, while the RESET process is attributed to the thermal dissolution of the metal filament due to Joule heating followed by the ion diffusion under concentration gradient and the applied electric field. Temperature measurements of switching behaviors can be explained by classical nucleation theory, suggesting the validity of our proposed mechanism [5].

We succeeded to observe the ionic current associated with redox reactions at Cu(Ag)/Ta_2O_5 interfaces by means of cyclic voltammetry (CV) measurements [6]. The results clearly showed repeated oxidation and reduction reactions of Cu and Ag at the interface under voltage sweeping, as shown in Fig. 1. The concentration of generated ions and their diffusion coefficient were estimated from CV curves with different voltage sweep rates. The diffusion coefficient was much higher than that evaluated for Cu ions in thicker Ta_2O_5 films. It was also found that the redox current is enhanced and the forming voltage is reduced when the film density of Ta_2O_5 is decreased, which are realized by different deposition methods. This indicates that Cu and Ag are more oxidized at the interface with a more porous Ta_2O_5

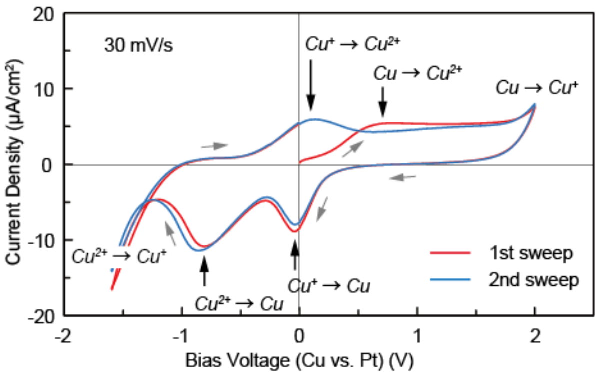

Fig. 1 Cyclic voltammogram of a Cu/Ta_2O_5/Pt atomic switch

layer.

Oxide films formed by any physical vapor deposition method typically have nanoporous structures that can absorb moisture from the ambient atmosphere. We found that the operating voltage varies as the ambient pressure level decreases, with the variation depending on the kind of oxide material used in fabrication [7]. The observed variation is attributed to moisture absorption into the oxide layer, which forms a hydrogen-bond network in the amorphous matrix [8]. To reveal the impact of moisture absorption, the redox reaction was investigated under controlled relative humidity (RH) conditions [9]. For a lower-density Ta_2O_5 layer, the ion concentration increased with increased RH, suggesting enhanced redox reactions by water uptake in the Ta_2O_5 layer. The corresponding diffusion coefficient decreased up to RH 50% due to ion-ion interactions, but increased when RH level was higher than 50%. This increased diffusion is considered to originate from proton conduction, because incorporated water molecules can dissociate into H^+ and OH^- under hydrated conditions. These behaviors depend strongly on the film density of Ta_2O_5 matrix

Among various properties of nonvolatile memories, the switching speed (time) is one of the most important parameters. We evaluated the SET and RESET times of Cu(Ag)/Ta_2O_5/Pt cells by transient current measurements under the application of a short voltage pulse [10,11]. The SET time decreased exponentially with increasing pulse amplitude, reaching as low as 1 ns using moderate pulse voltages. From a comparison with atomistic nucleation theory, the nucleation process is likely to the rate-limiting process for the SET operation. The RESET time decreased with increasing OFF resistance. The cell exhibited several ns for OFF resistances of ~600 Ω. The electrical power needed to RESET the cell is inversely proportional to the ON resistance, suggesting that the RESET operation involves thermal effects. These results also support our switching model.

3. Quantized conductance and synaptic behavior

In addition to usual bi-stable switching behavior, the unique characteristics of the atomic switch are conductance quantization and synaptic behavior. The conductance of Ag/Ta_2O_5/Pt cells was found to change in a stepwise fashion under constant small voltage bias (< 100 mV), which is attributed to the formation of an atomic point contact [12]. By applying positive and negative voltage pulses to the Ag electrode, the cell exhibited stepwise increases and decreases in the conductance, respectively. The conductance level could be controlled by varying the amplitude of input voltage pulses, as shown in Fig. 2. Moreover, when the interval time of consecutive input pulses was tuned, the cell showed short-term memory and long-term potentiation behavior similar to those of biological synapses. This synaptic behavior is difficult to achieve by ON/OFF bi-stable switch based on conventional CMOS transistors.

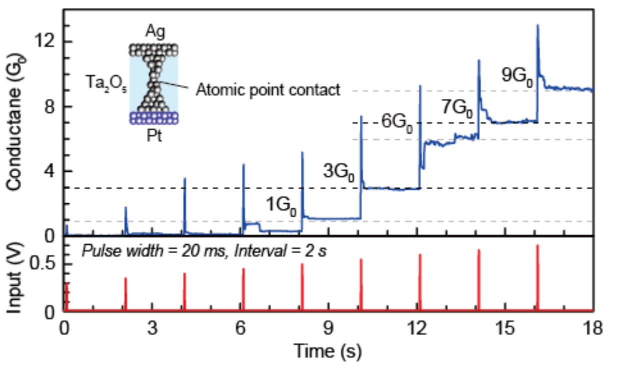

Fig. 2 Quantized conductance observed in an Ag/Ta_2O_5/Pt atomic switch

4. Conclusions

Atomic switches are new type of nanoionics devices that are operated by controlling ion transport and solid electrochemical reactions on the nanometer scale. The development of metal oxide-based atomic switches, which are fully compatible with the CMOS processes, has the potential for their commercialization. In addition to nonvolatile switching, new functions such as quantized conductance, learning ability, and synaptic behavior have been demonstrated, which can be used for neuromorphic applications.

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