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A Light-Controlled Resistive Switching Memory

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Resistive switching memory devices are systems in which the resistance of a material can be modulated between two non-volatile states by applying an electrical pulse. These devices are some of the most promising candidates for the next generation of non-volatile computer memories, although they are also being studied for many other advanced applications such as bio-inspired neuromorphic systems. Here we demonstrate a metal-insulator-semiconductor resistive switching memory in which the resistance can be controlled both by voltage pulses and by light. Our device stems from the traditional electrical-pulse manipulated resistive memories, adding the light as an extra control parameter. The system allows data encoding by using a certain light irradiance during the writing process and can be involved in the storage and transport of secured information. In addition, our design has the capability to work as light sensor, combination that opens the door to novel multifunctional storage devices.

There is currently an intense effort to push further the frontier of non-volatile storage devices. Resistive switching memories (RSM) are one of the most promising candidates to replace the current technology of non-volatile memories. The resistive switching in certain memory architectures was shown to be connected to the concept of memristors (memory resistors), initially introduced as a specific implementation of the fourth basic element present in an electronic circuit.[1,2] Resistive switching, typically measured in nanometric-sized metal-insulator-metal (MIM) structures, refers to the capability of switching an electrical insulator between a high resistance (HR) and a low resistance (LR) state by electrical pulses, allowing information to be stored as either 0 or 1.[3–6] The RSM systems are also emerging as key hardware elements for implementing bio-inspired neuromorphic systems, since their behavior can mimic the synapses between neurons.[7] Combined electric and magnetic field control of the memory state was reported in manganites.[8] In this paper we propose a new concept: a photon-charge bifunctional RSM with a simple metal-insulator-semiconductor (MIS) structure in which we can modify the resistance state not only by voltage pulses, but also by means of light. Our devices inherit the properties of the voltage or current controlled RMS, while adding the light as extra control parameter. To our knowledge, this is the first report on controlling the state of a resistive memory in a different manner, by using light in combination with voltage pulses.

The studied devices are vertical stacks with a metal/Al_2O_3 bilayer grown on a SiO_2/Si wafer (**Figure 1**). The fabrication involves the deposition of a 20 nm thick Al_2O_3 film on a p-doped epitaxial (100) Si substrate covered with a native SiO_2 layer, followed by a photolithography patterned Palladium layer (see Experimental Section for details). The Al_2O_3 films were prepared at 300 °C by Atomic Layer Deposition. The circular Pd top contacts have a radius of 50 μm and around each contact we defined rings with areas A_L from 10^{-5} cm^2 to 1 cm^2. These rings are left uncovered by metal, allowing light to reach the

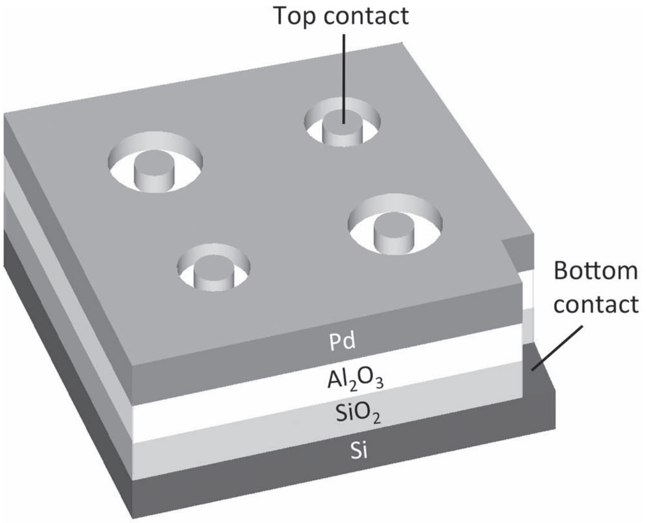

Figure 1. Schematic representation of the studied devices. An Al_2O_3 film is deposited on the SiO_2/Si substrate, followed by the fabrication of Pd top contacts. Around the circular Pd contacts (radius 50 μm) light-exposed Al_2O_3/SiO_2/Si rings are kept, with outer radii ranging from 53 μm to 1 cm, corresponding to ring areas, A_L, from 10^{-5} cm^2 to 1 cm^2. The remaining surface is covered with Pd to prevent light being transmitted to the optically active Si. The bottom electrical contact is on the p-doped Si substrate.

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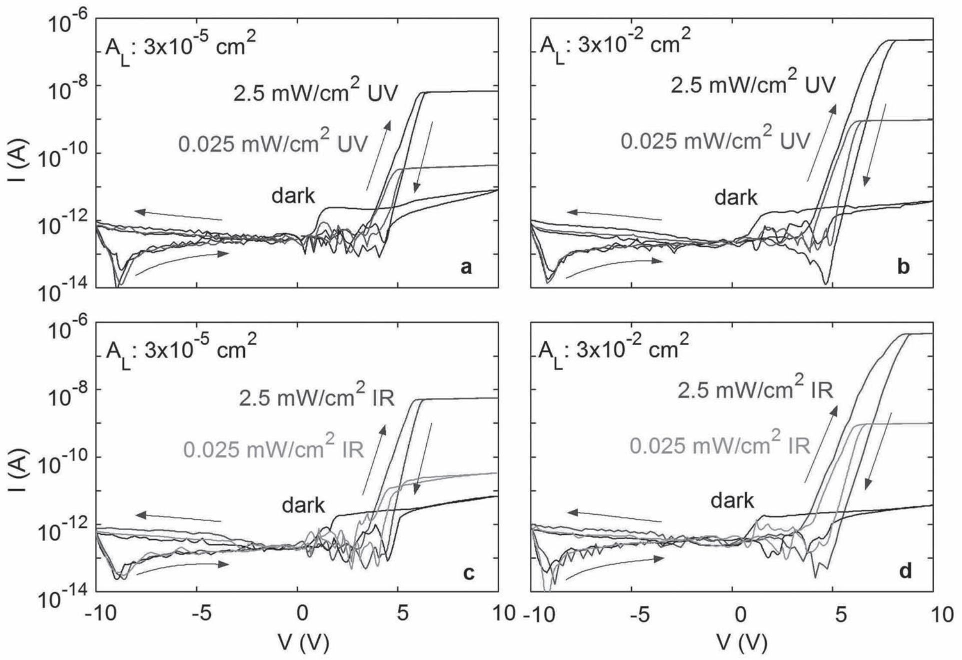

Figure 2. Typical current-voltage curves. *I–V* characteristics for a Pd/Al_2O_3/SiO_2/Si device upon different irradiances are presented (a,b - UV, c,d - IR) and light-exposed ring areas (A_L). The absolute values of the current are represented. The arrows indicate the memory operation sequence: a voltage of −10 V is equivalent to setting a low resistance state, while under positive bias the system switches from LR to HR state when the voltage exceeds a threshold value that depends on the illumination power. Consecutively measured *I–V* curves in the same conditions are superposed, so for the sake of clarity we only include one representative curve for each sample and irradiance value.

optically active Si after passing through the transparent oxide layers. The remaining surface was covered with Pd to block light irradiation.

The system we propose displays a light-dependent RSM behaviour in the ultraviolet (UV) - infrared (IR) range, and allows information to be encoded by using a chosen irradiance during the writing process. The observed dependence between the output current and the illumination conditions makes our design also suited for light sensing applications, thus allowing for the integration of an extra functionality in the non-volatile memory system.

The devices were investigated in dark conditions, as well as under illumination with UV (390 nm) and IR (950 nm) light emitting diodes (LED) with irradiances up to 2.5 mW/cm^2. In **Figure 2** we present the typical *I–V* curves measured at room temperature for two devices with different light-exposed Al_2O_3/SiO_2/Si ring areas, A_L (see Experimental Section for details). In what follows we concentrate on the *I–V* characteristics under illumination. Under forward bias, the system is initially in a LR state, while a voltage above a critical value switches it to a HR state. The HR state has the onset value shifted towards positive voltages with respect to the LR curve and hence, for a reading voltage of about +6 V, we observe a significant difference in the output current values for the two states. For reverse bias, which corresponds to a negative voltage applied to the Pd top contact, only a small current is measured independent of the illumination conditions. The *I–V* curves evidence a rectifying junction, with a blocking behaviour at reverse bias until the reverse breakdown is reached at about −15 V.

Under illumination, the *I–V* curves display a current saturation that reflects the photo-generated current in Si[9] (see Figure 2). By plotting the saturation current (defined as the current reached at +10 V) as a function of A, we observe a scaling up to an area of 0.03 cm^2; at this value the current reaches a plateau (**Figure 3**a). This saturation point corresponds to a ring radius of about 950 µm, which agrees with the electron diffusion length in the lightly-doped p-Si substrate.[10] Also, we note that under illumination with 1550 nm (0.8 eV) light, which is not sufficient for photogenerated electrons to overcome the Si bandgap of about 1.1 eV, at any LED power we only obtain the dark current. This result also proves that the heating produced by the LED is not influencing our devices. The photo-generated electrons in Si are the dominant charge carriers through the multilayer system.

For information storage we exploit the hysteresis present on the current-voltage curves in forward bias under illumination (Figure 2). The application of −10 V to the device is equivalent to bringing the system to a LR state (data writing), while +10 V are applied for switching to an HR state (data erasing). The reading pulse can be a chosen voltage between 4.5 V and 7 V. The memory window of the device can be tuned by changing the illumination and/or A_L. Consequently, we have extra degrees of freedom compared to conventional RSM.

The current through the device depends on the illumination conditions (Figure 3b). We can deduce the irradiance values by measuring the current for a known light-exposed area, A_L, at an appropriate applied voltage. This is equivalent to detecting in our system an extra capability as light sensor, offering an additional functionality to our memory devices.

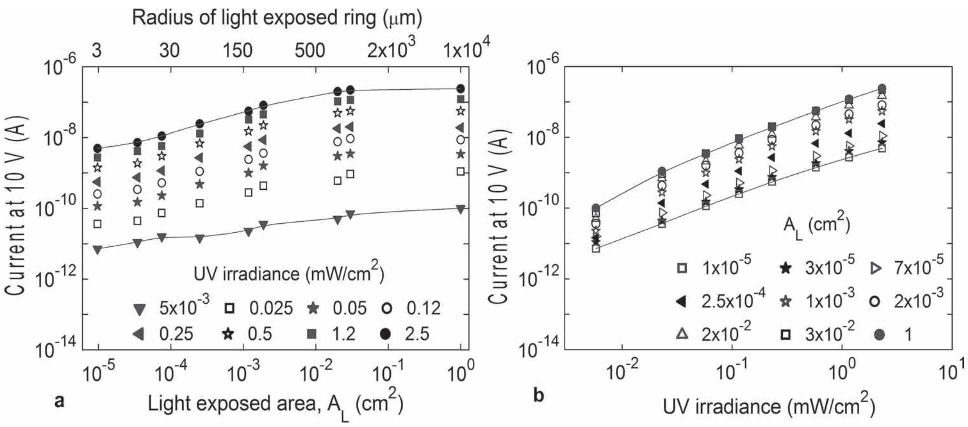

Figure 3. Light response. (a) Dependence of the current, measured at an applied voltage of +10 V, on the light-exposed ring area when illuminating the devices with UV light of different irradiances. The double abscissa scale helps correlating the light-exposed $Al_2O_3/SiO_2/Si$ ring radius and its corresponding ring area, A_L. (b) Dependence of the current, measured at an applied voltage of +10 V, on the irradiance of the UV LED for different light-exposed $Al_2O_3/SiO_2/Si$ ring areas around the Pd top contacts. These memory devices also work as a light detector, as proved by the strong relation between the output current and the irradiance.

Figure 4 presents bipolar hysteresis switching loops (HSL) measured for a device with A_L of 1 cm^2 under different illumination conditions. To construct the HSL, we apply voltage pulses of 5 ms following the sequence 0 V, +10 V, –10 V, 0 V, in steps of 0.1 V. After each of these pulse steps we wait 100 ms in short-circuit conditions, then we measure the remnant current (I_{rem}) for a fixed voltage of +6 V. This specific measurement of the remnant current reflects an authentic resistive memory behaviour, since capacitive effects (observed in Figure 2) are excluded by the long 100 ms time in short-circuit conditions.[11] In Figure 4, for certain regions of the HSL measured with 2.5 mW/cm^2 irradiance, we include sketches describing the trapped charges in the system.

We interpret the change in the remnant current as a modulation of the trapped electrons in the Al_2O_3 layer.[12,13] In this charge storage layer the resistivity increases upon the introduction of electrons from the Si layer.[14] For positive bias, a depletion region is generated in Si at the interface with the SiO_2/Al_2O_3. The photogenerated electrons are injected in the Al_2O_3 layer only if the electric field is high enough to overcome this energy barrier. In fact, in the HSL we observe an abrupt decrease of the remnant current at a voltage of about 7 V, which represents this effect. The charge removal occurs gradually at negative voltages (Figure 4). When the device is measured in dark conditions, the modulation in the remnant current is strongly suppressed, since the source of electrons is only the small dark current. We point out that a device with a thin SiO_2 layer alone does not present any hysteresis and there is no blocking at reverse bias, thus we ascribe the observed behaviour to the Al_2O_3 layer and the Al_2O_3/SiO_2 interface.

In order to verify the performance of the devices when used as core of a binary memory cell, we performed write/read/

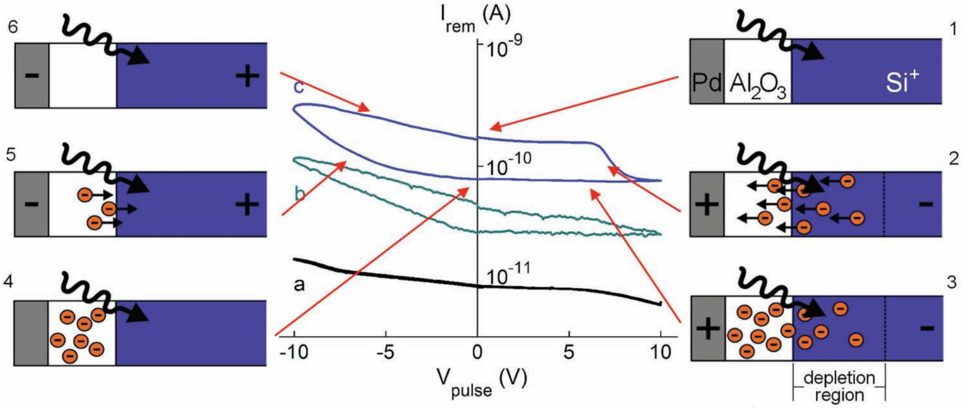

Figure 4. Remnant current hysteresis switching loops. The HSL were measured under different UV illumination conditions (a: dark, b: 0.025 mW/cm^2, c: 2.5 mW/cm^2) for a device with light exposed $Al_2O_3/SiO_2/Si$ ring area, A_L, of 1 cm^2. The remnant current was measured at 6 V considering that around this voltage value we have the best ratio between the LR and HR current values and the minimum noise in the high resistance state. For the HSL measured with 2.5 mW/cm^2 irradiance, we describe the behaviour of the trapped charges. Starting from LR at 0 V the Al_2O_3 layer is free of trapped charges (1). When the potential reaches 7 V, photogenerated electrons are injected in the Al_2O_3 layer (2), saturating the layer with charges and reaching the HR state (3). This state remains stable (4) until the potential exceeds a negative value that depends on the illumination conditions. (5), where the trapped charges are gradually removed from the Al_2O_3 layer (6).

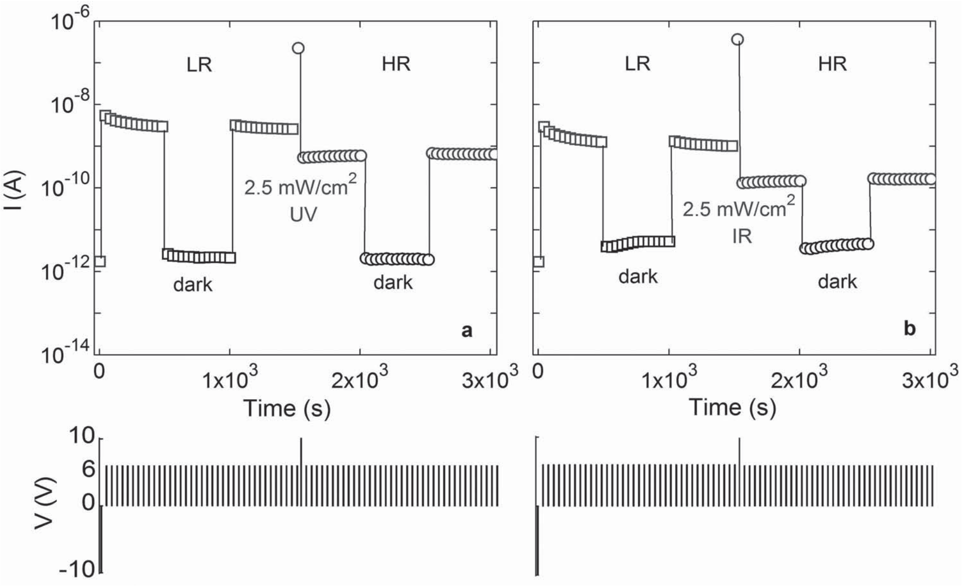

Figure 5. Data retention capability in dark conditions and under illumination with UV light (a), and IR light (b). Devices having light-exposed Al_2O_3/SiO_2/Si ring area, A_L, of 1 cm^2 were chosen in order to have a large memory window (see *I–V* curves in Figure 2). The voltage-pulses applied are displayed in the lower part of the figures. The ON (LR) state is set with one pulse of –10 V and consecutive readings at +6 V are made with light, in the dark, and then again with light. The OFF (HR) state was set with one pulse of +10 V and consecutive readings at +6 V were made with light, in dark, then with light.

erase/read experiments by applying 5 ms voltage pulses in the sequence –10 V/+6 V/+10 V/+6 V both in dark conditions and under illumination. We observe that data can be accurately written, read, and erased only under illumination (**Figure 5**). The light power during the writing step dictates an information encoding, the same light power being required for a correct read out of the data. Therefore, the devices can be employed to store and transport secured information. The storing conditions of the system between data processing steps do not influence the retention capabilities, and accurate readings are obtained both after maintaining the devices in the dark, as well as after keeping them under illumination.

The memory devices we propose were tested and support more than 10^4 write/read/erase/read cycles, with a data retention time, obtained by extrapolation, of about one year. We suspect that a gradual loss of the charges stored in the Al_2O_3 layer, probably due to electron diffusion, is responsible for the limited retention time.

We have designed a new type of resistive switching memory device with a simple metal/Al_2O_3/SiO_2/Si structure. This memory has extra capabilities when compared to conventional memory devices, as in our configuration information can be encoded by exposure to a certain light irradiance during the writing step. Furthermore, considering the dependence between the current and the light entering the system, the devices also function as light sensors. Multilevel switching can also be aimed at, considering that we have multiple steps for the LR state in the same device for different illumination conditions. As a final point, we note that the devices work with low currents, below the μA. This low-current operation is beneficial for memory devices, meaning fast data processing and reduced heat dissipation. Future efforts can be directed at improving the data retention time by tailoring the Al_2O_3 layer properties as well as reducing the device size. The results presented in this paper offer the possibility for extra degrees of freedom to be exploited in prospective complex memory architectures.

Experimental Section

The Al_2O_3 films were prepared at 300 °C by Atomic Layer Deposition (ALD) on single crystalline (100) Si substrates covered by a native SiO_2 layer. We used p-doped Si substrates (resistivity 15 Ω·cm) with an amorphous 1.9 nm thick native SiO_2 layer, as determined by ellipsometry. ALD involves sequential pulses of trimethylaluminium (TMA) as metal-carrying precursor gas and H_2O vapour as oxygen source, separated by N_2 inert gas purges. The deposition rate was 1.08 Å/cycle, with a time consumption of about 8 s/cycle. ALD provides atomically flat surfaces, supported in our case by the smooth native SiO_2 under-layer formed by oxidation in air of the single crystalline Si substrate. The lack of peaks in the grazing-incidence X-ray diffraction spectra for the Al_2O_3 films indicates an amorphous phase. The roughness of the Al_2O_3 surface was determined by X-ray reflectometry (XRR) to be below 0.2 nm, which means reduced optical losses from light scattering on surface imperfections when the device is illuminated. The Al_2O_3/SiO_2 bilayers are transparent to the incident light, which can reach the optically active silicon substrate. Photoelectrons are generated in the Si substrate under illumination. A 50 nm thick Pd layer was deposited by magnetron sputtering after photolithography on the Al_2O_3 film. Circular structures were created after lift-off (Figure 1).

The electrical characterization was performed at room temperature by current-voltage (*I–V*) measurements on a probe station connected to a Keithley 2636 source-meter. The p-doped Si substrate serves as grounded bottom electrode. A compliance current of 10^{-6} A was fixed for all measurements to protect the system from possible undesired high currents. The sweep rates (for Figure 2) were between 6 s/curve and 100 s/curve, similar results were obtained in all cases.

We used LEDs as light source, thus avoiding heat transfer to the sample, which was placed at a distance of 3 cm. The emitted UV and IR irradiation was calibrated using a commercial photodetector (Thorlabs DET36A).

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