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An experimental proof that resistance-switching memories are not memristors

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It has been suggested that *all* resistive-switching memories are memristors. The latter are hypothetical, ideal devices whose resistance, as originally formulated, depends *only* on the net charge that traverses them. Recently, an *unambiguous* test has been proposed [J. Phys. D: Appl. Phys. **52**, 01LT01 (2019)] to determine whether a given physical system is indeed a memristor or not. Here, we experimentally apply such a test to Cu-SiO_2-based electrochemical metallization memory cells, and show that electrochemical metallization memories are *not* memristors. Since the particular resistance-switching memories employed in our study share similar features with many other memory cells, our findings refute the claim that all resistance-switching memories are memristors. They also cast doubts on the existence of ideal memristors as actual physical devices that can be fabricated experimentally. Our results then lead us to formulate two *memristor impossibility conjectures* regarding the impossibility of building a model of physical resistance-switching memories based on the memristor model.

Introduction – Although some publications[1,2] have claimed that the memristor[3] (in the ideal sense) has been found and *all* resistance-switching memories are memristors[4], several researchers have raised serious doubts about such claims[5–10]. Indeed, the property of pinched hysteresis loops[11] alone ("If it's pinched it's a memristor"[12]) *cannot* serve as a good indicator of memristors since that property is shared by different types of experimentally-realizable devices (such as memristive devices and systems whose memory depends on some internal degrees of freedom, other than the charge[13]).

Remarkably, the most important characteristic of *any* memristor[3], namely, the functional dependence of its memory resistance (memristance), R_M, on *only* the net charge, q, that traverses it, $R_M(q)$, has *never been demonstrated experimentally*. Following the scientific method, it is obvious that any claim of the "memristor discovery" must be based on the *experimental* measurement of $R_M(q)$, and not merely on non-exclusive characteristics.

In Ref. 14, two of us (YVP and MD) have introduced a simple test to experimentally determine whether a resistor with memory is an ideal memristor or something else. The main idea of the test is based on the duality property of a capacitor-memristor circuit whereby, for any initial resistance state of the memristor and any form and amplitude of the applied voltage, the final state of an ideal memristor must be identical to its initial state, if the capacitor charge finally returns to its initial value[14]. In other words, our test verifies the $R_M(q)$ dependence.

To prove the ideality, a scan over a large range of parameters (initial state, shape/magnitude of the applied voltage, etc.) is clearly required. To prove the opposite, however, even a single measurement demonstrating the absence of such a duality would be enough.

In the present paper, we experimentally apply the ideal memristor test[14] to Cu-SiO_2 electrochemical metalization cells as model resistance-switching devices. Electrochemical metalization cells[15] constitute a large family of resistance-switching devices based on the cation diffusion through a solid electrolyte. Typically, such cells exhibit bipolar resistance switching with thresholds. This property allows us to use such Cu-based devices as representatives of the entire class of bipolar threshold-type resistance-switching cells[16].

Our experimental results show that the resistance-switching memories are *not* memristors, and cast further doubts on the existence of ideal memristors as actual physical devices that can be fabricated in the laboratory or found in Nature. This leads us to formulate two *memristor impossibility conjectures*, namely that *i*) it is impossible to accurately model physical resistance-switching memories by adding small corrections to the ideal memristor model, and *ii*) it is impossible to build a circuit combining *ideal* memristors with any other ideal two-terminal devices (resistors, capacitors, and inductors) that emulates realistically the response of experimentally-realizable resistance-switching memories.

Experimental details – Figure 1(a) shows the experimental circuit used to implement the ideal memristor test. To generate the test voltage signal $V(t)$ and control signal V_c we use a source measure unit (Keysight B2911A). The unit is controlled by a code written in C Sharp. To initialize and measure the device state, we close the relay (part number HI05-1A66, Standex-Meder Electronics), thus connecting the tested device to the source measure unit directly. To run the test, we open the relay and apply the test voltage signal (such as the triangular pulse in Fig. 1(b)) across the capacitor (non-polarized 10 μF capacitor) connected in series with the tested device.

The resistance-switching devices studied in this work were

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fabricated by sputtering deposition technique on the surface of a silicon wafer (substrate). A thin adhesion layer (5 nm Ti) was first formed on the surface of the substrate. We used 30 nm Ru as an inert bottom electrode common for all devices. A 10-nm-thick SiO_2 layer was deposited using a shadow mask with 10×10 mm square openings. The top Cu electrodes of 30 nm thickness were deposited on top of SiO_2 using another shadow mask with square and circular openings of various sizes (in this Letter we present data for a device with a circular top electrode of $r = 710$ μm). A 5 nm CoCrPt was used as a protective layer for the top electrodes (see inset of Fig. 2 for a schematic of the structure of memory cells). In order to dope SiO_2 with Cu atoms[17] the devices were subjected to 580 °C, 1 hour annealing in He environment. After that, the samples were slowly cooled down to the room temperature. The result is a typical resistance-switching memory cell with characteristics similar to many other experimental memory devices[16]. The ideal memristor test was implemented on several randomly selected devices showing stable switching behavior.

Results – Figure 2 shows typical current-voltage characteristics of a selected Cu-SiO_2 device. This plot demonstrates a bipolar switching with well defined thresholds, and a hysteresis loop twisted at the origin. From this plot we estimate the following parameters of our device: $R_{on} \simeq 19.5$ kΩ, $R_{off} \simeq 150$ kΩ, $V_{t,+} \simeq 0.7$ V, and $V_{t,-} \simeq -0.8$ V. Here, $R_{on/off}$ are the boundary resistance values and $V_{t,+/-}$ are the threshold voltages.

The ideal memristor test, as represented in Fig. 1, was performed at several values of the pulse amplitude V_0 (see Fig. 1(b)) with the initial memristance set to $R_M = 53$ kΩ. Here, we present the results obtained at $V_0 = 0.4$ V and $V_0 = 1$ V. Since these measurements were performed in sequence, the final state after the application of $V_0 = 0.4$ V served as the initial state for $V_0 = 1$ V. We emphasize that according to the test procedure, the initial and final charge on the capacitor is the same (zero). Therefore, if the tested device were a memristor, its final and initial memristance would be the same too.

It is found that at $V_0 = 0.4$ V the final memristance is the same as the initial one (cf. Figs. 3(a) and 3(c)). However, the larger value of $V_0 = 1$ V causes the device to switch into the

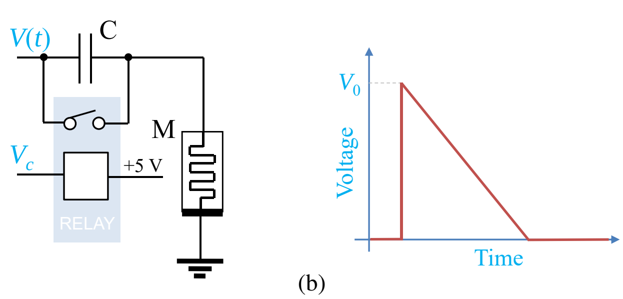

FIG. 1. (a) Capacitor-resistive memory circuit employed in our experiments. Here, a relay is used to shunt the capacitor, C, to initialize and read the state of the resistive memory, M. (b) Shape of the testing voltage $V(t)$ employed in the present work.

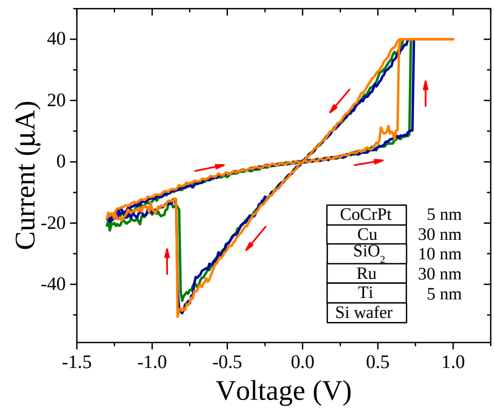

FIG. 2. Current-voltage characteristics of a Cu-SiO_2 device obtained using sweeps of 0 to +1 to -1.3 to 0 V with a positive current compliance of 40 μA, and a negative current compliance of 1 mA. Inset: Schematic diagram of the Cu-SiO_2 device structure.

lowest resistance state, $R_{on} = 19.5$ kΩ, see Fig. 4. As the final device state is *different* from the initial one when the capacitor has discharged, we conclude that *our device has not passed the ideal memristor test*. We observed similar results for all the other samples tested. Therefore, *none* of our devices have passed the ideal memristor test.

We note that the test was performed using 40 μA current compliance, which was not exceeded during the test. However, due to the nature of our test, its conclusions are *independent* of whether the current was limited or not by the compliance current.

Resistance-switching memories are definitely not memristors – We can further expand on these experimental results as follows. In this work we have applied the ideal memristor test suggested in Ref. 14 to Cu-SiO_2-based electrochemical metallization memory cells, which are a type of resistance-switching memories. As part of the test, we have compared the initial device state with the final one obtained under the condition of a capacitor discharge in series with the memory device. Since under at least one bias condition we have found that the final state of the memory devices was different from the initial one, we conclude that these resistance-switching devices cannot be described simply by a memristance that depends on the charge only: $R_M(q)$. Therefore, they are *not* memristors. Since the current-voltage characteristics of the devices used in our study are typical of a wide range of resistance-switching cells, our general conclusion is that *resistance-switching memories are not memristors*, irrespective of their specific device structures and switching mechanisms.

We also note that the triangular-shape voltage signal $V(t)$ employed in our work has facilitated the ideal memristor test. Under the test conditions of Fig. 1, the tested devices were subjected first to a relatively large positive voltage (the initial magnitude is V_0), followed by a small negative voltage. The tested devices failed the test since the positive voltage across the devices was sufficient to switch R_M to R_{on}, while the negative voltage was not sufficient for the inverse switching. Since,

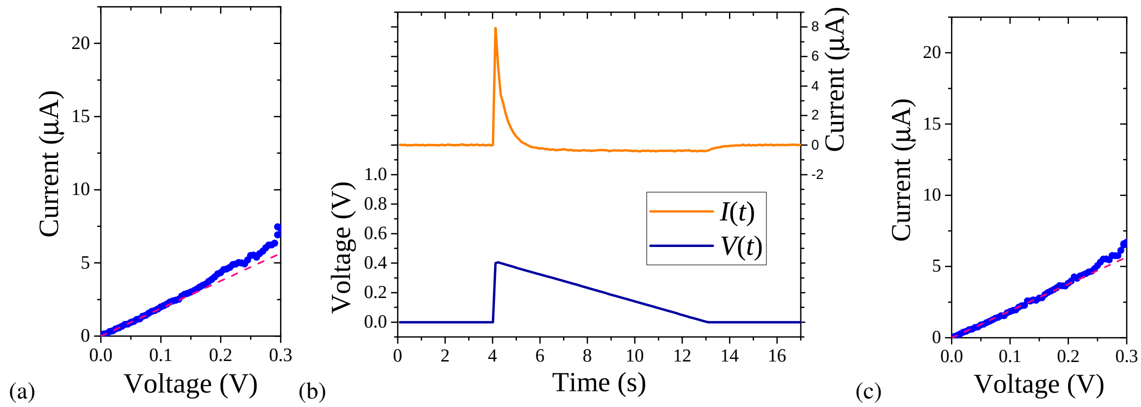

FIG. 3. Ideal memristor test performed at $V_0 = 0.4$ V. (a) A low-amplitude sweep is used to test the initial memristance (the relay is closed). (b) Voltage and current versus time, when the testing voltage is applied (the relay is open). (c) A low-amplitude sweep is used to test the final memristance (the relay is closed). The fitting lines in (a) and (c) correspond to the same $R_M = 53$ kΩ.

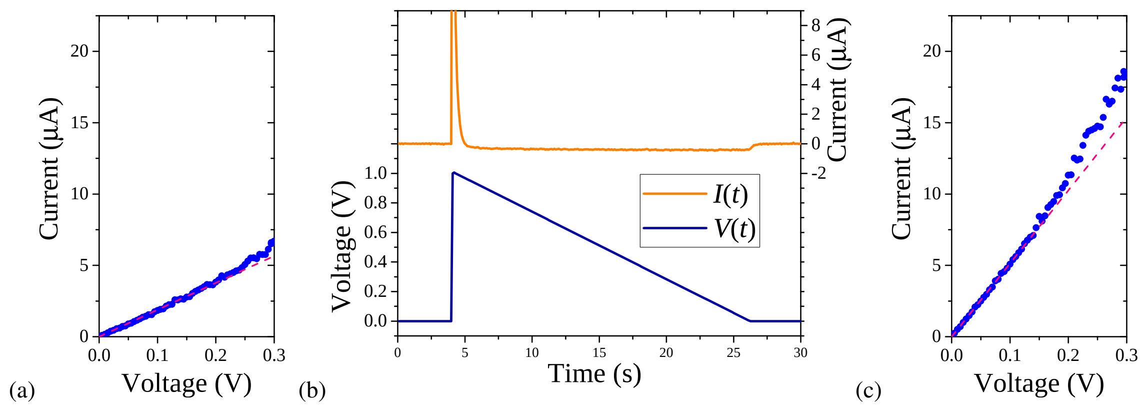

FIG. 4. Ideal memristor test performed at $V_0 = 1$ V. (a) A low-amplitude sweep is used to test the initial memristance (the relay is closed). (b) Voltage and current versus time, when the testing voltage is applied (the relay is open). (c) A low-amplitude sweep is used to test the final memristance (the relay is closed). The fitting line in (a) corresponds to $R_M = 53$ kΩ, while in (c) to $R_M = 19.5$ kΩ.

in principle, for any given pair of $V_{t,+}$ (positive voltage) and $V_{t,-}$ (negative voltage), one can always choose the test signal such that $V_0 > V_{t,+}$ and $V_M(t) > V_{t,-}$, one can further argue that *there are no ideal memristors among the threshold-type resistance-switching memories*.

Memristor impossibility conjectures – At this juncture, the reader may ask how the ideal memristor model is related, if at all, to physical resistance-switching devices such as those studied experimentally in this work, or any other similar devices published in the literature (see, e.g., references in 16). Accounting for the fact that the response of physical devices is different from the ideal behavior, Chua argued[4] that the resistance-switching memories are an “unfolding” theory extension of the ideal devices. More recently, he also proposed[18] that the resistance-switching can be represented by a circuit combining ideal memristors with some other ideal devices[19]. However, these statements are clearly *incorrect*.

In fact, unfolding relies on families of mathematical functions that are similar (close) to each other. When an idealized model is partially inadequate, the model can be improved by adding *small terms* resulting in the new model: an unfolding of the original system[20,21]. However, this approach is not applicable to the physical (experimentally-realizable) memristive devices because the difference between their physical models (as known in the literature) and the ideal memristor model can not be bridged by small correction terms. Similarly, a circuit representation of physical memory devices by circuits of ideal components is highly unlikely for the same reason: the ideal memristor behavior is too different from that of physical devices.

Based on the above arguments, we formulate two *memristor impossibility conjectures* that may serve as foundations for future research.

First memristor impossibility conjecture. It is impossible to accurately model physical resistance-switching memories by adding small corrections to the ideal memristor model.

Second memristor impossibility conjecture. It is impossible to accurately model physical resistance-switching memories by a circuit combining *ideal* memristors with any kinds of non-linear *ideal* circuit elements.

In the second conjecture, we refer to the ideal elements defined in Ref. 19. It can be also formulated in the strong sense considering only the combinations of memristors with basic circuit elements (non-linear resistors, capacitors, and inductors).

Conclusions – In conclusion, we have employed a recently suggested test[14] to experimentally verify whether resistance-switching memories are indeed memristors, as it was claimed in Ref. 4, or not. Our results demonstrate *unambiguously* that *they are not*.

Unlike the behavior of ideal memristors, the final states of the memory devices we have measured significantly deviate from their initial states. This study led us to formulate two conjectures on the impossibility of building a model of physical (experimentally-realizable) resistance-switching memories based on the ideal memristor behavior. The collection of these experimental results cast further doubts on the existence of the ideal memristor as a forth circuit element that can be fabricated experimentally.

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