

Lithionic two-terminal resistive switching devices with analog memory and a nanobattery architecture based on $\text{Li}_{1-x}\text{CoO}_2$ cathode, SiO_x electrolyte and $\text{Li}_{4+3x}\text{Ti}_5\text{O}_{12}$ anode

P. S. Ioannou^a, E. Kyriakides^a, O. Schneegans^b and J. Giapintzakis^a

^a Complex Functional Materials Laboratory, Department of Mechanical and Manufacturing Engineering, University of Cyprus, 75 Kallipoleos Ave., 1678, Nicosia, Cyprus

^b Laboratoire de Génie Electrique et Electronique de Paris, CentraleSupélec, CNRS, Université Paris-Saclay, Sorbonne Université, 11 rue Joliot-Curie, 91192, Gif-sur-Yvette, France

Memristor technologies are emerging as the most viable successor to CMOS-based transistor technologies for the implementation of neuromorphic computing, due to their two-terminal design which allows the non-von Neumann architecting of bioinspired artificial neural networks (ANN's), enabling the reduction of the energy- and silicon “real-estate”- costs [1].

A new class of ion migration-based resistive switching (RS) devices, that rely on the electrochemical Li^+ migration to produce reversible changes in the device conductance, has shown potential as binary or analog memristors [2], and artificial synapses [3].

The insulator to metal transition of $\text{Li}_{1-x}\text{CoO}_2$ (LCO) during delithiation ($0 \leq x \leq 0.25$) has been utilized in thin film nanobattery devices based on LCO cathode, SiO_2 electrolyte and Si anode to produce multistate RS, through the reversible field-driven migration of Li^+ from the LCO cathode to the Si anode [4]. Recently, devices based on $\text{Li}_{4+3x}\text{Ti}_5\text{O}_{12}$ (LTO, $0 \leq x \leq 1$) with a Pt/LTO/Pt architecture have shown non-volatile RS with analog conductance modulation capabilities, through the electric field driven Li^+ migration which leads to a phase separation between insulating $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and metallic $\text{Li}_7\text{Ti}_5\text{O}_{12}$ domains and the formation of conductive bridges between the two Pt electrodes [5].

Leveraging on mutually-beneficial electric field driven delithiation and lithiation of $\text{Li}_{1-x}\text{CoO}_2$ cathode and $\text{Li}_{4+3x}\text{Ti}_5\text{O}_{12}$ anode respectively in terms of conductance tunability, and the “zero”-strain property of LTO during lithiation in terms of endurance enhancement, the fabrication of Au/LCO/ SiO_x /LTO/Pt RS nanobattery devices is hereby examined. Such devices are capable of non-filamentary, non-volatile, and analog RS with prolonged retention ($\sim 10^5\text{s}$) and endurance ($\sim 10^4$ cycles). On the other hand, a small memory window ($I_{\text{on}}/I_{\text{off}} < 10$), strong potentiation/depression asymmetry, and indications of environmental-related degradation have been observed.

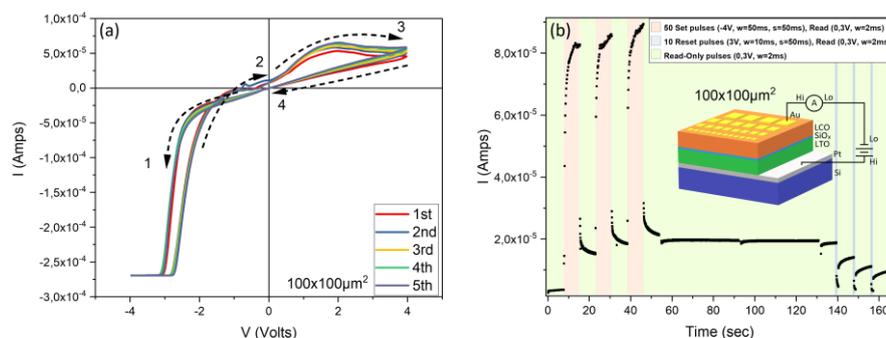


Figure 1: Current-Voltage characteristic curves of a typical Au/LCO/ SiO_x /LTO/Pt device, showing reversible RS (a). Pulsed voltage characterization of this device showing non-volatile multistate conductance modulation (b).

References:

- [1] Lanza, Mario, et al. "Memristive technologies for data storage, computation, encryption, and radio-frequency communication." *Science* 376.6597 (2022)
- [2] Zhu, Yuntong, et al. "Lithium-film ceramics for solid-state lithionic devices." *Nature Reviews Materials* 6.4 (2021): 313-331.
- [3] Ioannou, Panagiotis S., et al. "Evidence of biorealistic synaptic behavior in diffusive Li-based two-terminal resistive switching devices." *Scientific reports* 10.1 (2020): 1-10.
- [4] Mai, V. H., et al. "Memristive and neuromorphic behavior in a Li_xCoO_2 nanobattery." *Scientific reports* 5.1 (2015): 1-6.
- [5] Gonzalez-Rosillo, Juan Carlos, et al. "Lithium-battery anode gains additional functionality for neuromorphic computing through metal-insulator phase separation." *Advanced Materials* 32.9 (2020): 1907465.