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14. ABSTRACT

15. SUBJECT TERMS

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RPPR Final Report
as of 23-May-2023

Agency Code: 21XD

Proposal Number: 72621EM

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Report Date: 14-Oct-2022

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Final Report for Period Beginning 15-Jul-2019 and Ending 14-Jul-2022

Title: Plasticity Mechanisms and Neuromorphic Learning with Correlated Semiconductors

Begin Performance Period: 15-Jul-2019

End Performance Period: 14-Jul-2022

Report Term: 0-Other

Submitted By: Shriram Ramanathan

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees:

STEM Participants:

Major Goals: Major goals of the project include understanding fundamental mechanisms that govern electronic plasticity in model correlated semiconductor systems and implement the dynamic features in neural learning algorithms.

Accomplishments: We have studied non-associative learning in nickel oxide (NiO) based devices under a variety of external stimulus. Similar to biological organisms, habituation and sensitization learning plasticity has been investigated with the influence of electrical stimulus. Such learning behavior of NiO devices results from dynamic modulation of its defect and electronic structure. The results inspire new neural network learning algorithms that can incorporate the non-associative characteristics and show improvement in performance.

Training Opportunities: The training opportunities include setup and measurement of electrical transport properties of semiconductors, fabrication of semiconductor devices using lithography, developing algorithms for neural networks, preparing journal papers and technical networking among collaborators for detailed understanding of experimental observations.

Results Dissemination: The results from this study were disseminated through journal papers, presentations.

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Shriram Ramanathan

Person Months Worked: 1.00

Funding Support:

Project Contribution:

National Academy Member: N

RPPR Final Report
as of 23-May-2023

Participant Type: Co PD/PI

Participant: Kaushik Roy

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Sunbin Deng

Person Months Worked: 9.00

Project Contribution:

National Academy Member: N

Funding Support:

ARTICLES:

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Peer Reviewed: Y **Publication Status:** 1-Published

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Volume: 11

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Date Submitted: 7/6/20 12:00AM

Date Published: 5/1/20 4:00AM

Publication Location: London

Article Title: Perovskite neural trees

Authors: Hai-Tian Zhang, Tae Joon Park, Ivan A. Zaluzhnyy, Qi Wang, Shakti Nagnath Wadekar, Sukriti Manna,

Keywords: memory, correlated, trees, synapse

Abstract: Trees are used by animals, humans and machines to classify information and make decisions. Natural tree structures displayed by synapses of the brain involves potentiation and depression capable of branching and is essential for survival and learning. Demonstration of such features in synthetic matter is challenging due to the need to host a complex energy landscape capable of learning, memory and electrical interrogation. We report experimental realization of tree-like conductance states at room temperature in strongly correlated perovskite nickelates by modulating proton distribution under high speed electric pulses. This demonstration represents physical realization of ultrametric trees, a concept from number theory applied to the study of spin glasses in physics that inspired early neural network theory dating almost forty years ago. We apply the tree-like memory features in spiking neural networks to demonstrate high fidelity object recognition, and in future can open new directio

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Date Submitted: 5/19/23 12:00AM

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Article Title: Selective area doping for Mott neuromorphic electronics

Authors: Sunbin Deng, Haoming Yu, Tae Joon Park, A. N. M. Nafiul Islam, Sukriti Manna, Alexandre Pofelski, Qi

Keywords: phase transition, correlated, electronics

Abstract: The cointegration of artificial neuronal and synaptic devices with homotypic materials and structures can greatly simplify the fabrication of neuromorphic hardware. We demonstrate experimental realization of vanadium dioxide (VO₂) artificial neurons and synapses on the same substrate through selective area carrier doping. By locally configuring pairs of catalytic and inert electrodes that enable nanoscale control over carrier density, volatility or nonvolatility can be appropriately assigned to each two-terminal Mott memory device per lithographic design, and both neuron- and synapse-like devices are successfully integrated on a single chip. Feedforward excitation and inhibition neural motifs are demonstrated at hardware level, followed by simulation of network-level handwritten digit and fashion product recognition tasks with experimental characteristics. Spatially selective electron doping opens up previously unidentified avenues for integration of emerging correlated semiconductors

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Date Submitted: 5/19/23 12:00AM

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Publication Location:

Article Title: All-Electric Nonassociative Learning in Nickel Oxide

Authors: Sandip Mondal, Zhen Zhang, A. N. M. Nafiul Islam, Robert Andrawis, Sampath Gamage, Neda Alsadat .

Keywords: learning, Mott, NiO

Abstract: Habituation and sensitization represent nonassociative learning mechanisms in both non-neural and neural organisms. They are essential for a range of functions from survival to adaptation in dynamic environments. Design of hardware for neuroinspired computing strives to emulate such features driven by electric bias and can also be incorporated into neural network algorithms. Herein, cellular-like learning in oxygen-deficient NiO_x devices is demonstrated. Both habituation learning and sensitization response can be achieved in a single device by simply controlling the magnitude of the electric field. Spontaneous memory relaxations and dynamic redistribution of oxygen vacancies under electric bias enable such learning behavior of NiO_x under sequential training. These characteristics in simple device arrays are implemented to learn alphabets as well as demonstrate simulated algorithmic use cases in digit recognition. Transition metal oxides with carefully prepared defect concentrations can

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RPPR Final Report
as of 23-May-2023

Partners

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I certify that the information in the report is complete and accurate:

Signature: shriram ramanathan

Signature Date: 5/19/23 11:44AM

Final report

Contract number: W911NF-19-2-0237

PI: Shriram Ramanathan, Purdue University

Program Manager: Dr. Joe Qiu

Abstract: We have studied non-associative learning in nickel oxide (NiO) based devices under a variety of external stimulus. Similar to biological organisms, habituation and sensitization learning plasticity has been investigated with the influence of electrical stimulus. Such learning behavior of NiO devices results from dynamic modulation of its defect and electronic structure. The results inspire new neural network learning algorithms that can incorporate the non-associative characteristics and a brief summary of results are discussed below.

Non-associative learning emulation in NiO devices:

Semiconductor synthesis: NiO film was grown from a pure Ni target, using magnetron sputtering with a power of 100 W, where the applied voltage and current were controlled to 350 V and 300 mA (direct current) respectively. The NiO film was grown at a pressure of 5 mtorr with flowing of 1 sccm O₂ (2%) and 49 sccm (98 %) Ar gas mixture. The film was grown at a temperature of 300 °C with a rate of ~ 3 nm/min. To achieve the uniform film, the substrate rotated with a speed of 20 rpm during the deposition. A schematic of typical two-terminal metal-oxide-metal (MOM) device architecture is depicted in Figure 1. NiO is deposited by magnetron sputtering method on Ti/Pt (10/70 nm) coated silicon substrate (p-Si, 100). The silicon wafer was heated to 200 °C to remove the moisture from the surface before depositing the Ti/Pt metal. The bottom metal contact (Ti/Pt) was grown by the e-beam evaporation. Then the wafer transferred into the deposition chamber to grow 96 nm NiO film. After the growth of NiO film, the substrate was transferred to another sputtering chamber with a circular shadow mask to deposit the top metal electrode. A 200 nm palladium (Pd) top electrode was deposited by sputtering at 10–3 mbar pressure.

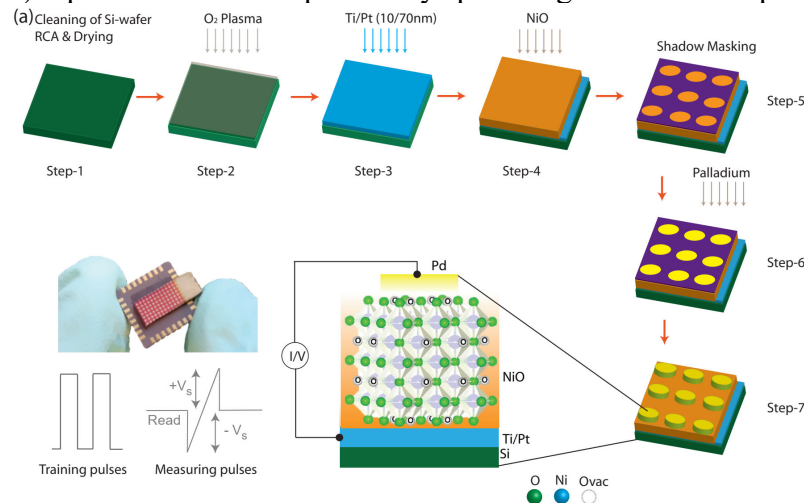


Figure 1. Process steps of device fabrication. (a) In step-1, the silicon wafer was cleaned using RCA cleaning process and then dried using N₂ gas. Following that the surface of the wafer was further cleaned using the oxygen plasma in step-2. The bottom electrode (Ti/Pt) was deposited on the silicon wafer by e-beam deposition system in step-3. Afterwards, the NiO film has been deposited through sputtering technique by covering a small part of bottom electrode (step-4). Further, NiO film is masked with metal shadow mask in step-5 and Palladium metal is sputtered on the NiO (step-6). The device is ready for the electrical measurement as shown in the step 7. (inset)

Electrical characterization: The electrical characterization of the solid-state NiO devices was performed by constantly measuring the current-voltage (I-V) curves by sweeping the gate voltage from -10 mV to 10 mV with a step of 5 mV using the Keithley 2635A source meter. The measurements were performed in a closed shield probe station to avoid electrical noise from the surroundings. The habituation and sensitization training in the array of devices have been performed by applying square pulses to the devices connecting them in parallel to the Keithley 2635A source meter. The NiO device shows a higher response to the training cycle and demonstrates a change in relative resistance of 7.6 % after the first training cycle. The relative resistance change reduces to 4.4 % and 3.8 % after the second and third training cycles respectively. The response of the NiO device gradually decreases with the repetition of the training cycle, indicating habituation to electric pulses. On the other hand, the response of the NiO continuously increases when the training pulses were applied with higher amplitude (V_1) of 30 mV.nm⁻¹ by keeping exactly identical pulse width and repetition interval as used for the habituation experiment. The early response of the device was found to be 13.6 % (first training cycle) and then decreased 8 % (second training cycle), indicating initial habituation, however, the response starts to increase after the second training cycle. The response of the devices become equivalent to the initial response (~13 %) after eight training cycles. A significant increase in the response to 19 % when the eleventh cycle of training pulses was applied to the devices.

Learning algorithm inspired by device behavior: Habituation has been shown to result from the decreased release of chemical transmitters at synaptic terminals, a stabilization mechanism known as synaptic plasticity. We previously demonstrated the usefulness of adding habituation-based learning to spike-timing dependent plasticity (STDP) in order to perform lifelong learning without catastrophic forgetting, and we connected that behavior with proposed oxide devices. More recently, we explored the addition of sensitization learning. In addition to dynamics that require the habituated gradual modulation of plasticity there may also be sudden changes in the environment, such as the introduction of novel information, which require a sudden and temporarily large increase in plasticity in order to avoid global corruption between old information and new information. In biology, this plasticity modulation (such as the sensitization behavior observed in the species *Aplysia*) is in part aided by neuromodulators, such as dopamine, which can be released when novel information is encountered. And, just as dopamine is released locally in the brain by dopaminergic neurons, the described sensitized response in neural networks should be local, targeted at specific reference vector neuron(s) that can be “reassigned” to the novel information with minimal loss of older information to prevent catastrophic forgetting. We note a similarity between this important learning function and the observed ability to stimulate the Mott insulator NiO to sensitize its resistivity change response to environmental exposure, even after a period of habituation. We have designed and implemented dopaminergic learning similar to this material functionality, showing the possibility of incrementally learning new information under rapid environmental changes without catastrophically forgetting useful, older information. Figure 2 shows the benefit of using this sensitization behavior in such a dynamic environment. Classes are presented sequentially, one after the other, without revisiting samples from previously seen classes. The sensitization behavior is targeted at less-used neurons that then rapidly learn novel data, which is identified by poor recognition by any reference vector neurons. The targeted sensitization allows for the untargeted neurons to retain their useful positions as centers of previously seen clusters. The materials and neuroscience driven algorithm described above have to be implemented in a programmable hardware architecture for efficient learning. The synaptic

memory will use the proposed devices in a memory array so that efficient reading and writing is possible for perception and training, respectively. We developed non-linear circuits to interface with the proposed devices to mimic computation primitives of deep spiking neural networks. The neural circuit primitives were assembled to form a spatial hardware architecture. Further work on system level implications of such an architecture with the proposed synaptic devices is currently under way.

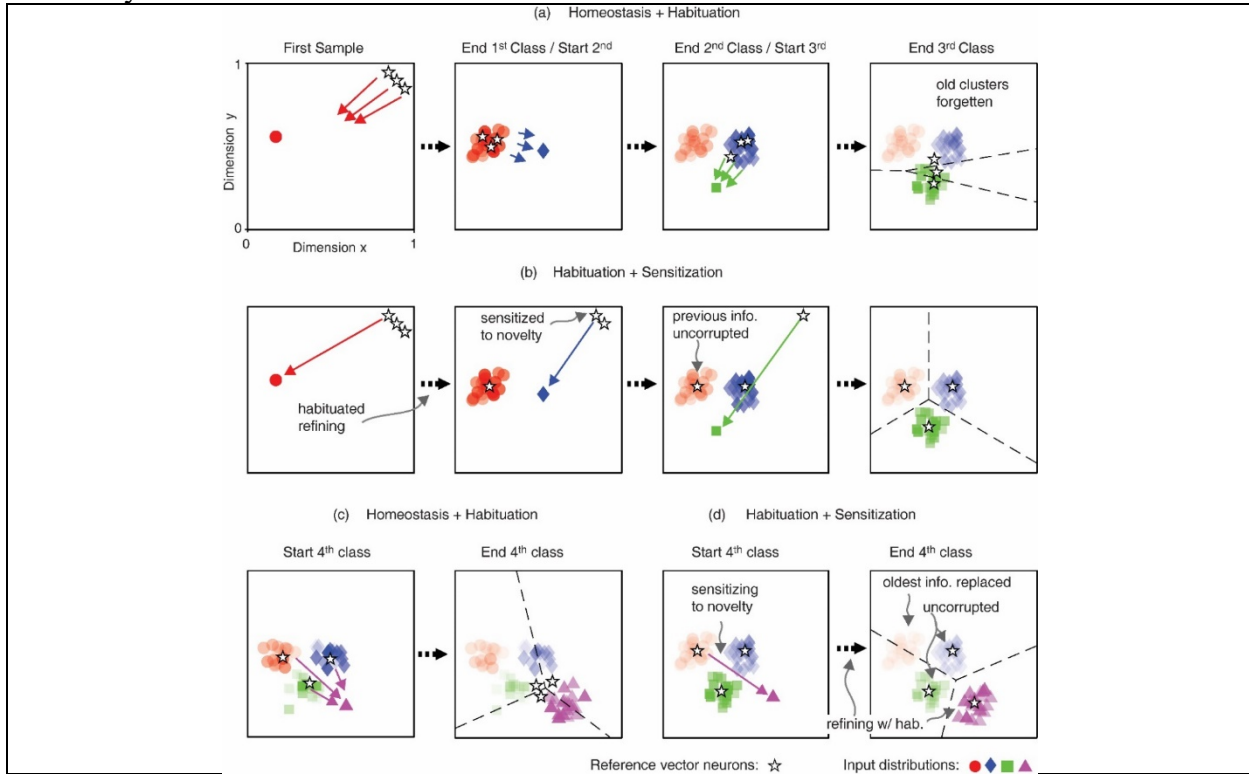


Figure 2: Snapshots of simulations of sequentially-presented classes (red, then blue, then green) without revisiting previously seen classes. In clustered learning, reference vector neurons seek to migrate toward the centers of the various data distributions. In (a), homeostasis with habituation—which typically operates correctly when data classes are interleaved—fails to retain centers on the older distributions. In (b), the use of sensitization to stimulate responses by less-used neurons in the presence of poor input detection (novelty) instead of traditional homeostasis allows for the already-trained neurons to be uncorrupted by the new data. Subfigures (c) and (d) show the same learning mechanisms as (a) and (b), respectively, but when attempting to learn a new, fourth class (magenta) in a memory-constrained situation after being pretrained on the three original classes. The addition of sensitization allows for selective forgetting of the oldest class without catastrophically forgetting the other classes.

Transient learning via leaky characteristics: The leaky characteristics of NiO devices have been tested by applying the voltage pulse and measuring the corresponding change in resistance (Fig. 3). Before applying the pulse, the device demonstrates a resistance state of 2.48 k Ω . A square voltage pulse of +1V with pulse width of 1s was applied to the devices. The device demonstrates resistive switching characteristics by reaching a new resistive state of 2.58 k Ω . This resistive state was continuously measured for 150s, and we observed the decay of resistance state with respect to time. Further, a negative voltage pulse of amplitude of -1V/1s was applied to the device and resistances were measured. The resistances of the device were switched to lower resistive state of 2.42 k Ω and relaxed back to original resistive state after 150s. Such relaxation behavior is also

correlated with forgetting behaviors of NiO. The resistance of the device was continuously measured by applying by sweeping the gate voltage of ± 10 mV.

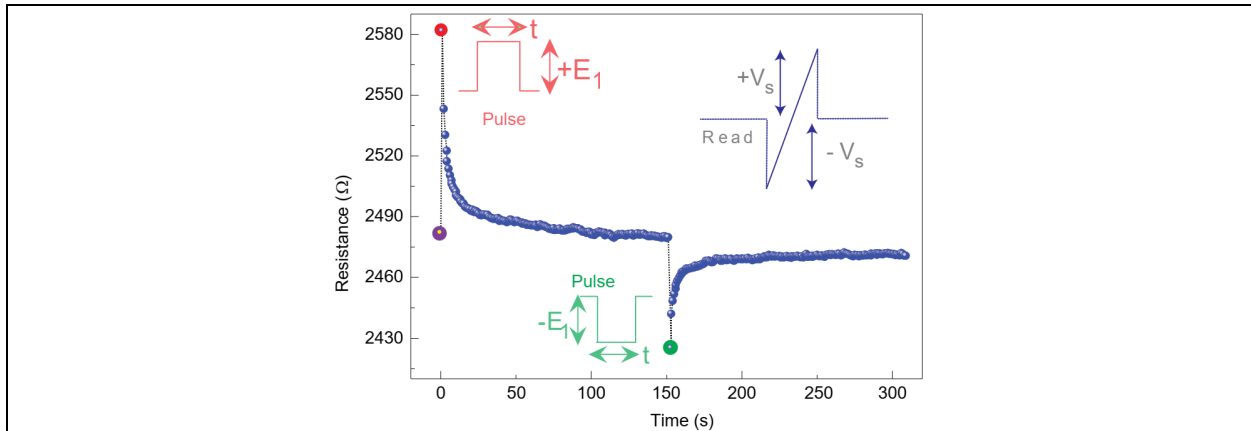


Figure 3. Relaxation of resistance state after applying different magnitudes of pulses. (Inset) Red and green square pulse for $+1$ V and -1 V respectively. The pulse with of the applied pulses is 1s. The triangular pulses are applied to measure the in-situ resistance state of the devices.

The relaxation behavior was studied over few cycles. Initially, positive amplitude of pulses ($+1$ V/1s) was applied to the device for four cycles (one pulse on each cycle) with an interval of 125s (Figure 4). The resistance of the device was measured during the interval and observed a relaxation of resistance to the original state of the device. Further the negative pulses were applied, and lower resistive switching characteristics were recorded. A similar relaxation characteristic was observed in the devices. However, the highest resistance measured for each pulse (2.60 k Ω , 2.59 k Ω , 2.58 k Ω and 2.58 k Ω) were slightly lower while consecutive pulse measurement was performed. The device slowly got habituated with respect to the electric field hence lowering of resistance switching characteristic.

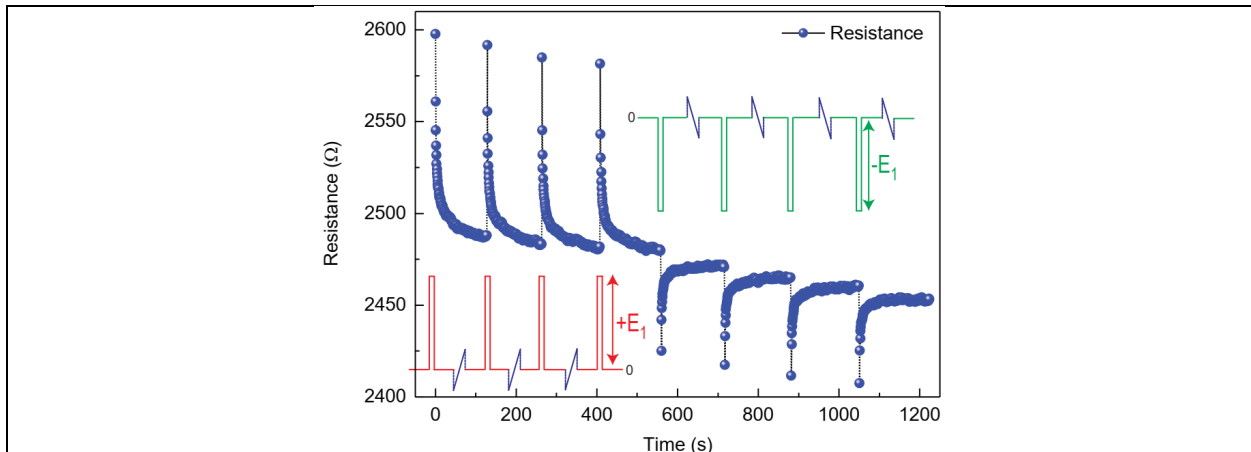
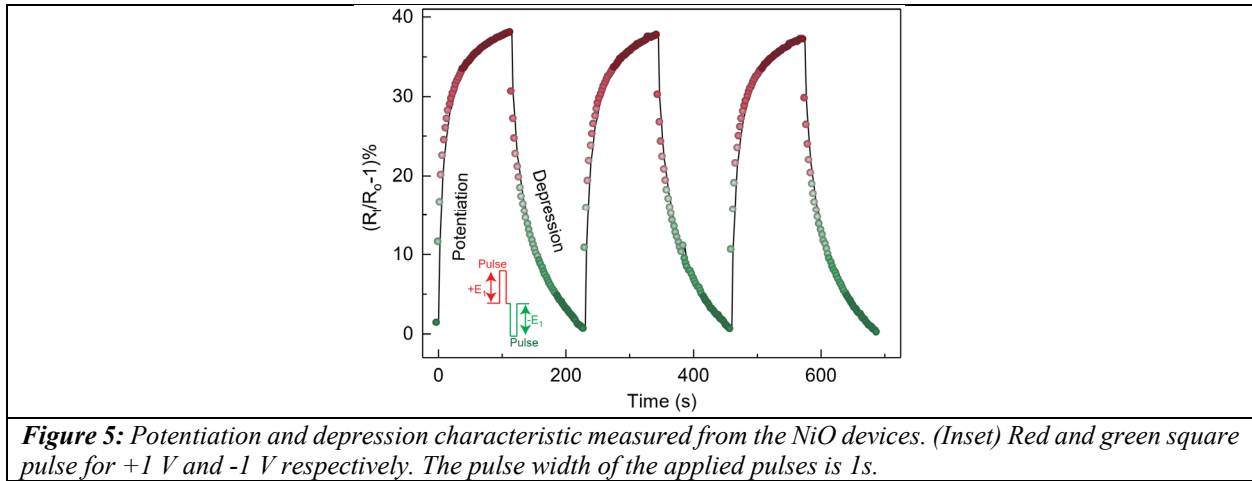


Figure 4: Resistive relaxation characteristics of NiO devices under multi-pulse operation. (Inset) $+E_1$ and $-E_1$ pulses are $+1$ V and -1 V respectively. The pulse width is 1s. The triangular pulses are applied to measure the instantaneous change in resistance state of devices

Among the most fundamental characteristics of learning and forgetting are potentiation and depression respectively. The potentiation measurement was performed by continuously applying 50 square voltage pulses of amplitude +1V with width 1s (Figure 5). The electric pulses were applied at an interval of 1.3 s. A strong change in relative resistance state $[(R_t/R_o - 1)\%]$ of 38% was measured. Such increase in resistance state with respect to applied voltage pulses are attributed to potentiation characteristics of devices. Similarly, the depression characteristic has been observed while negative voltage pulses were applied to the device. The negative voltage pulses help to decrease in resistance state and back to the original state. Continuous potentiation and depression measurement was performed over few cycles. The device demonstrated reproducible characteristics without any degradation.



Summary: In summary, we have presented representative results from NiO devices on electrically controlled resistance switching and gradual decay. Phase transitions from a correlated state into a conducting state due to migration of charge carriers enable transient memory formation. Such short-term memory can be utilized for emulating various learning behavior in neural networks and complement algorithmic implementation of learning. Emerging semiconductors such as correlated oxides are potentially relevant for neuromorphic computing and artificial intelligence.