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Inducing alternating nanoscale rectification in a dielectric material for bidirectional-trigger artificial synapses

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Nanoionic device-based artificial neural networks that consume little power and hold a potential for enormous densities still fall behind the capabilities of software algorithms running on traditional von Neumann machines. In addition, despite many publications showing multilevel parametric capabilities associated with these devices, a real-world nonvolatile memory application that maximizes their potential density is yet to be realized. One reason may be due to their limited functional mode as an analog passive element that is crippled by large interdevice variations. This work demonstrates that the nanoscale stoichiometry in transition metal oxides can be triggered to form asymmetric cationlike vacancy distributions that yield dynamically toggled current rectifying properties. In this manner, a rectifying device operated as an artificial synapse is capable of switching between excitatory and inhibitory modes, dissipating ~ 20 fJ/switching event. This complementary functionality (in a similar manner to CMOS transistors) adds a whole new degree of freedom to the design of neuromorphic computing platforms. Moreover, the entire spectrum of nonvolatile states derived from different cation distributions (positive-rectifying, negative-rectifying, conductive, and insulating) may be considered as a mutually exclusive and interchangeable basis set for robust multilevel memory implementation that overcomes the issues associated with large process and device related parametric distributions. *Published by the AVS.*

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I. INTRODUCTION

Nanoionic devices^{1–3} have been extensively used for the implementation of artificial neural networks in brain-inspired computational schemes⁴ with crossbar array architectures being promising candidates for achieving high densities ($\sim 10^{15}$ bits/cm²) similar to the human cerebral cortex ($\sim 10^{15}$ synapses), especially when configured in 3D stacking.^{5–8} These architectures are very efficient in terms of calculation time and energy expenditure when performing matrix operations.⁹ Array-based networks were demonstrated of being able to accomplish tasks such as image classification,¹⁰ starting from small scales of only few neurons,¹¹ medium levels^{12,13} up to hundreds of neurons,¹⁴ and even 10^5 synaptic connections.¹⁵

A chemical synapse is a gapped connection between two neurons through which communication takes place.^{16–19} A typical neuron can have several thousands of synapses that mostly connect axons in a presynaptic neuron to dendrites in a postsynaptic neuron. Interneural signaling occurs by the release of neurotransmitters from the presynaptic neuron into the gap (i.e., synaptic cleft), which in turn are collected by receptors in the postsynaptic neuron. The postsynaptic response is classified as being either excitatory or inhibitory and determined by the type of neurotransmitter used during signaling. Two key characteristics resulting from this behavior are the long-term potentiation (LTP) and long-term depression (LTD), where the synaptic weight (the connection

strength) is modulated by a neural activity. A recent study showed that two types of neurotransmitters could be released simultaneously during the synaptic activity.²⁰ Moreover, neurotransmitters have been shown to be able to actually exchange roles under certain circumstances.²¹

Artificial synaptic devices aim to imitate as much as possible the features that biological synapses have and in particular the ability to adjust the weight parameter as a function of the activity. To successfully reproduce this process, these devices should provide a high degree of freedom in the form of multilevel operation. Yet, a direct correlation exists between device dimensions and the number of states it can support (i.e., multilevel resistance).²² Unfortunately, sizing up increases the overall current and power consumption as well as reduces the prospective density. A desirable weight parameter (e.g., analog conductance) should be both symmetric with regard to the direction of change (increase or decrease) and differentially linear in terms of magnitude per step-change. Nonetheless, redox-based and other types of memristors show nonlinear conductance as well as asymmetric conductivity changes^{23,24} in response to successive set and reset pulses.^{25–27}

The ability to reconfigure dynamically between the excitatory and inhibitory modes, in a similar manner to a biologic synapse, is yet another desirable trait. Main stream nanoionic devices lack the ability to reproduce this feature since they are passive by nature (once formed) and operated to comply with the linearity requirement as much as possible. Conventional resistive random access memory (RRAM) devices are, thus, unable to display this dynamic attribute without adding an additional control terminal to modulate

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the material properties by the field effect. Additionally, it dictates the use of exotic materials as the resistive switching medium. Furthermore, adding a control terminal would both damage the linearity and seriously downgrade the high-integration capabilities. Recent publications have demonstrated devices with dynamic capabilities (mimicking either LTP or LTD behaviors as a function of a modulation bias) through the use of FET structures.^{28–32} These devices are operated using very large biases (tens of volts due to the physical structure and the material used) that hinder their integration possibility with modern CMOS applications. Moreover, their physical dimensions are very large (tens of micrometers) in addition to having a lateral structure that is unsuited for 3D integration.

Self-rectification devices were initially designed to target the “sneak-path” issue in large scale arrays.³³ In this approach, a complementary structure was constructed by mirrored layering of thin film materials that effectively implemented metal cation diffusion-based resistive switching. However, apart from having a relatively complicated structure, the underlying directional rectification mechanism presents no viable option to reset the device to its initial zero current insulating state. Other concepts exploit the nonlinear materials’ switching characteristics³⁴ to incorporate a self-rectification element into the device operation, but the characteristics do not present good enough results for the operation of large crossbar array applications, in addition to other issues such as CMOS incompatibility. Self-rectifying schemes that resolved the compatibility and structural complexity issues were also suggested, but only with static and unidirectional rectification abilities.^{35,36}

II. EXPERIMENT

A. Experimental setup and methodology

A schematic illustration of the measurement system setup is given in Fig. 1. Electrical measurements were performed at 300 °K using a conductive atomic force microscope (CAFM) in ultrahigh vacuum (UHV) of $\sim 10^{-10}$ Torr pressure and 15 mV set-point force in the static contact mode. CAFMs were widely used in the study of the resistive switching effect.^{37,38} The CAFM probe material is Si coated with a polycrystalline boron-doped diamond and is electrically connected to an external Keithley SCS4200 semiconductor parameter analyzer. The CAFM probe functioned as the first electrode, and the Au second electrode was grounded through the mounting stage as illustrated in Fig. 1. The

FIG. 1. Schematic diagram of the CAFM-based experimental setup.

nanosize contact area between the probe and the hafnia surface allowed the latter’s resistance to be modulated within a very confined region using a voltage sweep supplied by the parameter analyzer to the probe. Precise modulation of the resistance was accomplished by a current compliance (CC) setting in the parameter analyzer, which capped the current flow to a preset value through automatic adjustment of the applied voltage. The scan speed was tuned to compromise between the need for accuracy (slow scan) and minimization of thermal drift (fast scan). In order to obtain image resolutions of at least 64–128 lines taken over an area of about $40 \times 40 \text{ nm}^2$, an acquisition time of roughly 2 min was used, which gives a scan rate of $\sim 20\text{--}40 \text{ nm/s}$.

B. Material preparation

First, a blanket sample of $\sim 20 \text{ nm}$ uncapped hafnia (HfO_x) thin film was fabricated on a p-Si substrate. The hafnia was formed by atomic layer deposition using tetrakis (dimethylamino) hafnium as the metal precursor and H_2O vapor as the oxidizer. The growth temperature and pressure were 250°C and 0.1 Torr, respectively, without postdeposition annealing. Second, a lithographic process was used to pattern $1 \mu\text{m}$ wide rectangular structures over the oxide followed by a de-scum process and reactive ion etching of 5 nm into the hafnia to create a shallow trench. A gold thin film layer of 100 nm was then evaporated followed by lift-off process and diluted isopropyl alcohol (IPA) cleaning. Finally, the samples were baked at 110°C for 10 min.

C. Structural analysis

The x-ray photoelectron spectroscopy (XPS) characterization of the deposited hafnia 20 nm thick thin film revealed a metal to oxygen ratio of $\sim 1:1.6$ as presented in Fig. 2.

III. RESULTS AND DISCUSSION

A. Interchangeable mutually exclusive memory states

The conceptual idea of a bidirectional-trigger rectification device is depicted in Fig. 3. The illustration shows the cyclic evolution between four cation-vacancy configurations that form nonvolatile memory states in a resistive switching layer (RSL) redox material. The RSL (transparent) is confined between two conductive electrodes (top and bottom planes) that are biased either with a positive voltage polarity or with a negative one. As a result, the induced electric field leads to the generation and migration of defect states composed of positively charged oxygen vacancies (OVs) and interstitial negatively charged oxygen species (O).^{39–41} Given enough time, those current conducting defects align along the electric field lines, and a conductive filament (CF) takes shape. Electrons (e^-) flow through the CF along these vacancies, as if hopping over stepping stones, across the insulating RSL. Terminating this process, by resetting the bias to zero, after an initial conductive stub has formed but before a full CF was able to develop would result in a Schottky-like tunneling barrier next to one of the electrodes (either top or bottom depending

FIG. 2. XPS spectra of the hafnia sample showing the intensity core-level spectra for (a) Hf 4f and (b) O 1s. The stoichiometry ratio results in a slightly oxygen deficient HfO_x ($x \approx 1.6$) film.

FIG. 3. Schematic illustration of four interchangeable mutually exclusive states in a cation-vacancy-based nanoionic device (O represents negatively charged oxygen species, OV represents positively charged oxygen vacancies, and e^- represents electrons while the hopping current is shown by black directional arrows). (a) Negative-rectifying: a negatively biased forming process yielding in a tunneling gap (indicated by a dashed arrow) at the bottom with negative current rectification properties; (b) conductive: alleviating the current compliance restriction leads to a full body filament with symmetric current conduction represented by bidirectional arrows; (c) insulating: application of a more negative bias resulting in O-OV annihilation and zero current; (d) positive-rectifying: flipping the bias condition to a positive polarity along with restoration of the CC restriction induces migration of OV toward the bottom and positive current rectification due to the formation of a new tunneling gap (indicated by a dashed arrow) near the top.

on the bias polarity). This arrested forming may be viewed as a directional-trigger to generate a rectification state.

The first nonvolatile state is depicted in Fig. 3(a). Cation-OV defects are attracted to the negatively charged top electrode, while O anions are driven toward the bottom electrode. The forming process is stopped using a CC limitation. Once the current level through the RSL hits this preset limit, the bias is reset to zero and the forming process is stopped. This trigger setting results in an uneven defect distribution with a tunneling gap near the bottom electrode. Defect states located at the interface between the top electrode and the RSL have higher electron trapping probabilities (compared to defects located inside the RSL) due to wave function overlap. This, in turn, leads to a preferable direction for the electron flow from top to bottom (*negative-rectifying*, NR).

Reapplying the same bias while relaxing the CC limitation leads to the formation of a fully developed CF between the electrodes as shown in Fig. 3(b). The OV defect distribution is more symmetric with regard to the electrodes, and the tunneling gap disappears. As a result, electron hopping has no preferred direction, and current conduction between the electrodes is symmetric. This situation is defined as the second nonvolatile state (*conductive*). Increasing the bias at this point will lead to a higher current and Joule heating. The added heat encourages the recombination of interstitial O with OV defects, which leads to rupture of the CF and loss of the conducting ability. Figure 3(c) depicts the case where only a few scarce OV concentrations were left in the RSL leading to the third nonvolatile state (*insulating*).

The fourth nonvolatile state presented in Fig. 3(d) depicts a reversed asymmetric distribution of defects with the tunneling gap located near the top electrode. In this case, an opposite biasing scheme is used with a positive polarity applied to the top electrode. The electric field across the RSL is now in the opposite direction, and OV-cations redistribute to form a higher density near the bottom electrode. Once again the CC determines the positive trigger that yields a state with a preferable direction for the electron flow from bottom to top (*positive-rectifying*, PR).

B. Lateral test structure

A room temperature dynamic characterization of vacancy-based cations in a transition metal oxide in a traditional vertical structure is an extremely complicated task even for high end systems. An alternative approach, used in this work, takes advantage of the current conductive properties of these defects. The observational evidence for the underlying mechanisms that yield dynamic current rectification was accomplished through the use of CAFM for mapping and manipulation of defects, which were formed in a lateral arrangement close to the surface of the RSL. For this purpose, a lateral structure was fabricated over which the local nanoscale stoichiometry manipulation was done.

Figure 4 depicts a schematic diagram representing this structure for illustrative purposes. The structure is composed of an RSL adjacent to a grounded conductive gold electrode. A CAFM boron-doped diamond probe serves as the second

Fig. 4. Schematic depiction of a lateral device having a conductive stub and a Schottky-like tunneling barrier between the CAFM probe and Au electrode. The illustration represents a negative rectification state with regard to the Au electrode. A conductive AFM is used to manipulate and characterize the nanoscale stoichiometry in a transition metal oxide by electrical redistribution of cationlike vacancies along the surface to effectively produce a rectifying bidirectional-trigger memory device. It is shown that asymmetric densities result in mutually exclusive and interchangeable nonvolatile states with dynamic current rectification properties that could be utilized as a bilingual (excitatory and inhibitory) synaptic response (note: scale bar 5 nm).

electrode and was used for defect creation and annihilation by placing it in very close proximity to the Au electrode and inducing the electric stress. Defect mapping was later done through surface current scanning to study the effects of this stress. The gold electrode was placed in a shallow trench of ~ 5 nm to allow the confinement of electric field lines within the RSL while kept close to the surface. In order to reduce AFM probe convolution artifacts, slanted sections in the Au-RSL interface were chosen during the experiments. This, in turn, allowed for more reliable current mapping in close proximity to the electrode. The triggered forming of a partial conductive filament was done using a CC setting in a semiconductor parameter analyzer. Figure 4, thus, illustrates a negative triggered process where positively charged OVs were pushed away from the anode (Au) and accumulated near the cathode (probe), while the negatively charged O ions were drawn to the anode. The forming process is arrested by the CC setting before a continuous CF from the anode to the cathode is able to form and with the resulting gap yielding a current-rectifying state.

A slow scan in the contact mode was used along the surface of the sample while measuring the current through the probe and recording a 2D current map using a very low bias of about 10 mV. When the probe touched highly isolating oxide regions, literally zero current passed through the system. On the other hand, low current levels were measured when the probe came into close vicinity to trap rich regions. The lateral Au electrode was kept grounded, which resulted in a current overshoot when the probe came into contact with it or with a conducting filament-like region that effectively provided a current path to ground. In addition, very slanted regions along the lateral electrode were chosen to conduct the experiment in order to minimize the convolutional artifact.

C. Stoichiometry manipulation by OV redistribution

The first approach used in this work to dynamically trigger the current rectification is based on the manipulation

of defects by the electric stress induced spatial redistribution (Fig. 5). Initial triggered formation and the manipulation of OV-cations were done in consecutive steps, using a gradually increasing CC limit, followed by surface current map scanning. Each trigger step was conducted with the CAFM probe being placed over the same predefined location (indicated by a star in the figures). Figures 5(a), 5(c), 5(e), 5(g), and 5(i) show the consecutive I-V sweeps performed with alternating voltage polarities and CC conditions where defect formation incidents are marked by solid arrows. The quantification of the current rectification capability for a given voltage V_r was done by estimating the overall symmetry in the measured I-V curve. For this purpose, $I(V_r) \cong I(-V_r)$ is regarded as nonrectifying, while an asymmetric property where $I(V_r) \gg I(-V_r)$ (and *vice versa*) is considered as rectifying. Figures 5(b), 5(d), 5(g), 5(h), and 5(j) are the corresponding subsequent current mappings executed after each sweep. The images indicate the existence of two partially formed CF stubs with a primary one on the left, marked by white arrows in Fig. 5(b). The asymmetric distribution of cations in the main CF was initially higher near the probe as seen in Fig. 5(b) and after an additional negative sweep, given in Fig. 5(c), grew even thicker. This thicker CF is shown in Fig. 5(d). Curve 1 in Fig. 5(e) is yet another negative sweep that resulted in further thickening, seen in Fig. 5(f) (white arrow). Curves 2 and 3 in Fig. 5(e) exhibit NR properties with a rectification ratio (RR) of $I_2(-V_r)/I_3(V_r) \cong 20$ for $V_r = 0.5$ V. The voltage polarity of the successive sweep, curve 1 in Fig. 5(g), was switched to a positive one, and the CC increased to $1 \mu\text{A}$. It resulted in triggering an inverse special distribution of OVs as seen in Fig. 5(h). Traps migrated away from the probe and closer to the Au electrode. The rectification state was flipped to a PR with a ratio of $I_2(V_r)/I_3(-V_r) \cong 10$ for $V_r = 0.5$ V as evident in curves 2 and 3 of Fig. 5(g).

The final voltage sweep, given in Fig. 5(i), was done once again using a negative polarity and resulted in symmetric conduction ($RR \cong 3.5$ for $V_r = 0.25$ V in curves 2 and 3) due to the formation of a near complete CF as seen in Fig. 5(j). This result may be associated with surface phenomena related to the lateral structure. O anions can easily undergo desorption to the UHV environment given the electric stress and local Joule heating. Under these circumstances, it would not be possible to reset the device since no substantial O-OV recombination can take place. However, conventional lateral structures that are obvious candidates for real-world applications should show more robust behavior due to the confinement and reuse of O species within the material.

D. Stoichiometry manipulation by OV annihilation

A second approach used to dynamically trigger the current rectification is based on the manipulation of defects through O-OV recombination as illustrated in Fig. 6. This experiment was performed over a different location and conducted in a similar manner to the previous one. The current mappings are shown in Figs. 6(a)–6(c), while the corresponding voltage sweeps are given in Figs. 6(d) and

FIG. 5. Bidirectional rectification toggling by stoichiometry manipulation through OV redistribution (probe approximate location is marked by a star). (a) Initial formation by a negative I-V sweep (trigger events are marked by solid arrows). (b) Subsequent current mapping revealing newly formed OV near the probe (marked by white arrows), while some OV migrated away from the Au electrode (marked by a blue/shaded arrow). (c) A second negative I-V sweep induced OV annihilation (dashed arrow) and further OV forming (solid arrow). (d) A second current mapping showing CF thickening; (e) negative triggering by a third sweep with a CC of 100 nA resulted in NR. (f) A third current mapping revealing an asymmetric distribution of OV-cations having a higher density next to the probe. (g) Positive triggering by a positive sweep with a CC of 1 μ A resulted in flipping the rectification to PR. (h) Subsequent current mapping showing that OV traps redistributed and moved closer to the Au electrode. (i) A consecutive negative sweep resulted in nearly symmetric current conduction. (j) The final current mapping showing a fully developed, thick CF.

FIG. 6. Flipping of a PR state to an NR by OV annihilation (probe approximate location is marked by a star). (a) Initial current mapping. (b) Current mapping after the first sweep (positive trigger) showing a high trap traps near the Au electrode (indicated by a white arrow). (c) Current mapping after the negative sweep (negative trigger) revealing a redistribution of OV where recombination occurred near the Au electrode (indicated by a blue/shaded arrow). (d) An initial positive sweep using CC of 10 nA (forming is marked by solid arrows) and subsequent sweeps showing PR. (e) Negative sweep using CC of 10 nA (recombination events are marked by dashed arrows) and subsequent sweeps showing NR.

6(e) and marked as 1–6 according to their successive order. Figure 6(a) depicts the initial state that later evolved into a rectifying CF with a higher defect distribution near the electrode, as marked by a white arrow in Fig. 6(b). The initial positive voltage trigger, shown by curve 1 in Fig. 6(d), produced a CF with clear PR properties, as indicated by curves 2 and 3 in Fig. 6(d). The calculated RR for this state was 100 for a given $V_r = 0.5$ V. The following step was the application of a negative trigger followed by surface mapping. Curve 4 in Fig. 6(e) depicts a negative sweep with defect annihilation incidents (marked by dashed arrows), while the blue/shaded arrow in Fig. 6(c) shows the location where defects underwent recombination. In this case, less I-V sweeps were performed and the majority of O anions were not yet lost to the UHV environment, allowing for O-OV recombination. The PR state was, thus, flipped to NR, having a $RR \cong 10$ for $V_r = 0.5$ V as evident by curves 5 and 6 in Fig. 6(e).

E. Conduction mechanisms

The different current conduction mechanisms along the surface were investigated by placing the CAFM tip over different locations and registering the time-dependent current behavior. During this experiment, the tip was relocated alternatively between regions identified as metallic (Au), isolating (native RSL), or defect-rich, and the results are summarized in Fig. 7.

Initially, an additional positive sweep was performed over the same location, indicated by a star in Fig. 6(c), with a CC of 100 nA and followed by the mapping given in Fig. 7(a). The current mapping revealed newly formed OV defects (marked by a white arrow). At this point, the current was monitored as a function of time in a continuous manner while the AFM tip was placed over different locations on the surface marked by stars 1–8 in Fig. 7(a) (corresponding to the order of placement). The tip was initially placed over an isolating region 0, which showed current levels in the order of few picoampere. Moving the tip to the metallic regions (1, 3, and 5) resulted in current saturation (~ 10 nA due to the CAFM analog to digital converter range limitation). Once placed back on the isolating sections (2, 4, and 6), the current virtually dropped to zero. Figure 7(b) shows the current as a function of time where the numbers 1–8 on the graph correspond to the current level registered over the same placements of Fig. 7(a).

Finally, the tip was moved to locations 7 and 8 that are within 2 nm of the newly formed traps. In these positions, the time dependence clearly showed a stochastic nature, as expected from trapping and detrapping mechanisms that govern stepping stonelike electron hopping conduction. The average current level was about -2.5 nA as evident in Fig. 7(b). The temporal current in Fig. 7(c) (showing a zoom in over the time line between placements 7 and 8) reveals that the average current nearly doubles after the tip was relocated from 7 (roughly a distance of 2 nm from the traps) to 8 (within 1 nm of the

FIG. 7. Current conduction mechanisms for different locations over the surface. The CAFM tip was successively relocated (stars indicating the sequential placement of the tip) between the metallic and isolating regions (1–6) and eventually placed near the high-density OV region at locations (7 and 8). (a) A current map obtained after an additional positive sweep using CC of 100 nA reveals generation of new OV (marked by a white arrow). (b) A time-dependent current recorded for each point while the tip was successively moved along the surface. (c) A zoom-in showing the stochastic nature of the hoppinglike induced conductance through the traps.

traps). This behavior corresponds to the electrode-trap occupancy flux exponential model.

F. Theoretical analysis

Numerical simulations based on an analytical model³⁹ were performed to support the experimental results. Figure 8 summarizes the simulation results for OV and O densities (N_{OV} and N_O) in the RSL for the corresponding four mutually exclusive states shown in Fig. 3. Current rectification occurred mainly due to the existence of tunneling gaps near

the electrodes as shown in Fig. 8. Electron wave function overlaps along with higher quantum occupation probabilities for interfacial traps, making it easier for electrons to flow into the interfacial traps and tunnel through the gap than the other way around. For a detailed theoretical analysis of the interaction between O species and OV, including migration and recombination, the reader is advised to consult the provided references.

Figures 8(a) and 8(b) depict the cation and anion density distributions after the forming process was arrested at a CC condition of 100 nA while using a potential of -4.25 V on the top electrode (negative triggering). Figure 8(a) for N_{OV} indicates the existence of a hopping path through the RSL with a gap of about 0.5 nm near the bottom electrode (negative-rectifying). This gap corresponds to the tunneling barrier depicted in Fig. 3(a). The N_O density profile, shown in Fig. 8(b), is higher near the bottom electrode since the negatively charged O species were driven toward it by the electric field. As the process was allowed to proceed with a more relaxed CC limitation, a cation-rich conductive path was formed between the electrodes as seen in Fig. 8(c). Elimination of the tunneling barrier allowed for symmetric current conduction (conductive) corresponding to the state as shown in Fig. 3(b). The anion density in Fig. 8(d) near the bottom increased as well due to O generation and electric field driven migration.

At the next simulation step, the CC limitation was released even further, and Joule heating induced O-OV recombination leads to the dissipation of the conductive path resulting in zero current conduction (insulating). This unipolar reset phenomenon is known to occur with inert electrode materials. The state shown in Fig. 3(c) corresponds to the density profiles depicted in Figs. 8(e) and 8(f). Once the bias polarity of the top electrode was changed to $+4.25$ V and the CC limit restored to a level of 100 nA (positive triggering), the electric field driven migration flipped the cation distribution, and a tunneling gap of about 0.5 nm appeared near the top electrode (positive-rectifying). This reversed rectification state corresponds to Fig. 3(d), and the simulated density distributions are given in Figs. 8(g) and 8(h).

G. Discussion

Within the scheme of brain-inspired computing, memristors are commonly used in array architectures in a differential manner.⁴² In this approach, two passive devices form a single synaptic weight, thus allowing for both positive and negative parametric weight values. The synaptic weight-current is then evaluated through a differential amplifier to determine whether the synaptic gap represents an excitatory or an inhibitory state. Individual setting of the analog conductance for each of the two emulates the LTP and LTD synaptic behaviors. For the purpose of evaluating the synaptic weight, a rather complex differential amplifier sensing circuitry is needed. Network training of such differential architectures, when combined with an analog RRAM operation, requires nontrivial pulsing schemes that complicate the

FIG. 8. Simulated density behavior for OV traps and O species (N_{OV} and N_O) in the RSL showing four mutually exclusive states. Negative triggered formation using a negative bias (-4.25 V) with a CC of 100 nA, having a tunneling gap near the bottom electrode (negative-rectifying): (a) N_{OV} ; (b) N_O ; a fully developed CF formed by alleviating the CC conduction (conductive): (c) N_{OV} ; (d) N_O ; a more negative voltage bias applied to the top electrode resulted in CF dissipation and zero current conduction (insulating): (e) N_{OV} ; (f) N_O ; changing the bias polarity of the top electrode to a positive one ($+4.25$ V) with a CC of 100 nA resulted in positive triggered flipping of the rectification condition (positive-rectifying): (g) N_{OV} ; (h) N_O .

design of a digital controller especially for high density applications.

Alternating current rectification is an alternative approach used to imitate both excitatory and inhibitory synaptic functionalities.⁴³ Conductance-weight changes may be done by employing a digital strategy (similar to biologic synapses) where several devices are grouped together to represent a single artificial synapse.⁴⁴ Setting the number of current-rectifying units to a specific direction, either positive rectification or negative rectification, determines the overall conductance in each direction in a linear fashion that overcomes the nonlinearity issues associated with conventional devices. It allows dynamic switching of the rectification direction to account for either an excitatory or an inhibitory synaptic mode.

In terms of power dissipation, these devices consume very little power per synaptic event since they are based on the switching of a single CF. Given a switching current of 100 pA under an applied voltage in the order of ~ 2 V,⁴³ along with an upper limit of 10^{-4} s on the switching time as set by the measurement system sampling limitation (actual metal ion-based CFs were demonstrated to show switching times as low as few nanoseconds),⁷ the total energy dissipation

results in 20 fJ/switching event. This figure is not far from the 10 fJ associated with biological synapses and certainly much better than the capabilities of the micrometer sized devices mentioned in Introduction.

FIG. 9. Bidirectional-trigger rectifying nanoionic devices as a basis for array-based artificial binarized (± 1) neural networks have the potential to both increase the array density and simplify the design, training, operational complexity, and supporting peripheral circuitry by allowing dynamic switching of the rectification, thus implementing either an excitatory or an inhibitory weight parameter at each junction (like the CMOS couple shown in the inset).

Binarized neural networks⁴⁵ are artificial intelligence algorithms that utilize binary weight assignments during training and run-time. The weights in these networks are represented by a deterministic binarization (± 1) based on the assumption that the network can represent a full set of Boolean functions between all binary inputs and outputs. Binarized algorithms require less spatial complexity, memory bandwidth, and power consumption. It was also suggested that binary state neurons would greatly simplify hardware implementations.⁴⁶

A suggested hardware implementation for a binarized neural network based on bidirectional-triggered rectifying devices is shown in Fig. 9. It should be highlighted that this figure depicts a proposed crossbar array implementation for an artificial neural network for illustration purposes only and was not implemented in this work. This implementation allows the reader to better understand the advantages and flexibility embedded in a synaptic device based on the operation principles discussed herein.

The operation principle allows dynamic switching of the rectification direction resulting in either an excitatory or an inhibitory functionality of the artificial synaptic gap at each junction. In this manner, a single active device can be set to either push or pull current (much like the CMOS couple shown in the inset), thus simplifying the neural network design, training, operational complexity, and supporting peripheral circuitry overhead. The operation mode of traditional redox nanoionic devices in artificial neural networks is limited to a passive excitatory mode. In this scheme, the synaptic weight parameter corresponds to the conductance of the device, and weight adjustments are done using an analog approach. Inhibitory operations or negative weight implementations, thus, require creative architectural solutions that cost both resources and design complexity. In addition, the realization of real-world, high-density memory applications is inhibited by interdevice variations that cripple their multilevel capability when mass production is concerned. This study demonstrates that the nanoscale stoichiometry in transition metal oxides may be manipulated to form asymmetric cationlike vacancy distributions that yield bidirectional-triggered current-rectifying properties.

Based on the above discussion, a dynamically toggled rectifying artificial synaptic device would be capable of switching between excitatory and inhibitory modes. This complementary operation (much like CMOS transistors) adds a whole new degree of freedom for the design and operation of neuromorphic computing platforms. Moreover, the entire spectrum of nonvolatile states derived from different cation distributions (positive-rectifying, negative-rectifying, conductive, and insulating) may be considered as a mutually exclusive and interchangeable basis set for robust multilevel memory implementation that overcomes the large parametric distribution issues.

The dynamic nature of the rectification direction may be utilized for real-time modulation and adjustment of artificial synaptic plasticity. Since a prospective device, based on the phenomena discussed in this work, is essentially binary in nature, using a plurality of devices connected in parallel

would be needed to represent a single synaptic gap. By changing the ratio of the number of positive-rectifying to negative-rectifying individuals, the long-term plasticity may be controlled. This represents a digital approach (contrary to the common analog treatment) for implementing LTP and LTD synaptic weight adjustment.

IV. SUMMARY AND CONCLUSIONS

In summary, the operation mode of traditional redox nanoionic devices in artificial neural networks is limited to a passive excitatory mode. In this scheme, the synaptic weight parameter corresponds to the conductance of the device, and weight adjustments are done using an analog approach. Inhibitory operations or negative weight implementations, thus, require creative architectural solutions that cost both resources and design complexity. In addition, the realization of real-world, high-density memory applications is inhibited by interdevice variations that cripple their multilevel capability when mass production is concerned. This study demonstrates that the nanoscale stoichiometry in transition metal oxides may be manipulated to form asymmetric cationlike vacancy distributions that yield bidirectional-triggered current-rectifying properties. Based on that, a dynamically toggled rectifying artificial synaptic device would be capable of switching between excitatory and inhibitory modes. This complementary operation (much like CMOS transistors) adds a whole new degree of freedom for the design and operation of neuromorphic computing platforms. Moreover, the entire spectrum of nonvolatile states derived from different cation distributions (positive-rectifying, negative-rectifying, conductive, and insulating) may be considered as a mutually exclusive and interchangeable basis set for robust multilevel memory implementation that overcomes the large parametric distribution issues. The merits of using a proposed crossbar-structured, artificial neural network based on such theoretical devices were discussed as well.

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