MATERIALS SCIENCE

In-sensor reservoir computing for language learning via two-dimensional memristors

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The dynamic processing of optoelectronic signals carrying temporal and sequential information is critical to various machine learning applications including language processing and computer vision. Despite extensive efforts to emulate the visual cortex of human brain, large energy/time overhead and extra hardware costs are incurred by the physically separated sensing, memory, and processing units. The challenge is further intensified by the tedious training of conventional recurrent neural networks for edge deployment. Here, we report in-sensor reservoir computing for language learning. High dimensionality, nonlinearity, and fading memory for the in-sensor reservoir were achieved via two-dimensional memristors based on tin sulfide (SnS), uniquely having dual-type defect states associated with Sn and S vacancies. Our in-sensor reservoir computing demonstrates an accuracy of 91% to classify short sentences of language, thus shedding light on a low training cost and the real-time solution for processing temporal and sequential signals for machine learning applications at the edge.

INTRODUCTION

The massive amount of data produced and transmitted by the Internet of Things (IoT) requires new insight into real-time information processing by edge computing devices based on novel materials and architectures (1). Proactively interpreting and learning with temporal and sequential information represent key tasks for edge computing devices (2). However, current edge computing systems mostly rely on physically separated sensors and digital processing units, leading to high rate of energy consumption and long-time latencies when sequentially digitizing the analog pixel signals (3–9). The direct processing of time-varying optical data accounting for more than 80% of the collected information (10) or the perceptions of humans in a bio-plausible fashion would provide a breakthrough by reducing the communication and computation loads of edge computing devices that operate on the IoT.

While recurrent neural networks (RNNs) are highly capable of processing time-series data and various dynamic sequential events (11, 12), the high training complexity in RNNs limits their practical use with regard to edge computing on the IoT (2). Among various frameworks of RNNs, reservoir computing (RC) has demonstrated substantial reductions of the computational cost of learning, offering a promising solution by which to develop edge devices for temporal pattern classification, prediction, and generation (2). In RC, a dynamic reservoir is used to map complex inputs nonlinearly (e.g., spatiotemporal patterns) into high-dimensional states, which can be further augmented by virtual nodes, allowing a simple and fast training of readout weights at a low computation cost.

Thus far, RC systems have been physically implemented on conventional digital platforms and previously unidentified hardware dynamic systems powered by different physical mechanisms (e.g., electronic, photonic, spintronic, mechanical, and biological RC implementations) (6, 9, 13–18). For example, RC systems have been built on memristors with a simple structure, high density, good energy efficiency, and three-dimensional (3D) stackability (9). Memristors have demonstrated the ability to mimic the leaky integrate-and-fire operations of neurons in a manner that is more energy and area efficient than digital alternatives, as reported by Du et al., Moon et al., and Midya et al. (6, 8, 9). Conventional memristors mostly rely on redox reactions and the migration of ions and cannot directly respond to optical stimuli. Creating RC systems with high computing efficiency and direct responses to optical inputs without extra sensors/processors remains a challenge. Accordingly, memristors based on novel nanomaterials or hybrid materials (19–31) that offer different degrees of plasticity by means of either electrical or optical signals have been reported. However, the realization of an RC system with high computing efficiency enabled by in-memristor computing and direct responses to optical inputs without extra sensors/processors with a simple device structure remains a challenge.

In this study, we demonstrate optoelectronic RC for language learning with dynamic memristors built on a two-terminal tin sulfide (SnS) device structure. The synergy of the charge trap/detrapping dynamics and the photogating effects in the atomically thin material used in this study enables high-performance capabilities and versatile memristive behaviors with dual-mode operation (i.e., driven by electrical and optical stimuli), as well as good nonlinearity and fading memory. These memristors constitute an optoelectronic reservoir by responding to sequential electrical and broadband optical stimuli. Mapping complex temporal optoelectronic inputs into high-dimensional reservoir states, the optoelectronic RC described here demonstrates an accuracy of 91% in classifying practical Korean sentences with small natural errors. The low training cost and real-time processing of spatiotemporal signals for optoelectrical stimuli pave the way for efficient edge machine learning.
RESULTS
SnS memristor for optoelectronic RC

Optoelectronic RC for language learning is schematically illustrated in Fig. 1A, where optical stimuli representing a Korean sentence “사자” (“Let’s buy” in English) was directly used as input into a signal processing system without dedicated image sensors or associated analog-to-digital data conversion. The absence of extra optical sensors and data conversion, reducing the energy and time requirements considerably, represents the major benefit of the optoelectronic RC method beyond traditional RC (fig. S1). This faithfully resembles the biological neural system of photoreceptors in the retina (32). An RNN is embodied as complex connections between neurons (magenta circles), as shown in Fig. 1A, where the connections are not trained in the RC system. Instead, we train the connections of readout weights (colored arrows denoted by $\theta_i$ in Fig. 1A) linking the high-dimensional reservoir states and output neurons representing different sentences under the framework of supervised learning.

The key element of optoelectronic RC is its use of a dynamic memristor with dual-mode operations and fading memory with a 2D material, SnS (for which the Raman spectrum and transport curve are shown in fig. S2). Nonlinear fading memory with depression and facilitation features conductivity that can be modulated by electrical and optical stimuli, respectively, in the SnS (dual mode) (Fig. 1, B to E). This roots on the p-type SnS that has donor and acceptor states in its bandgap (i.e., gap states) originating from the atomic vacancies of Sn (acceptor) and S (donor) (33, 34). The schematic band diagram in Fig. 1F shows the equilibrium state of the p-type SnS for transport with immobile acceptor states occupied by electrons.

The decreasing current upon continuous current-voltage sweeps in Fig. 1 (B and C) and fig. S3 is unique among various 2D materials (fig. S4). The depression of conductivity can be explained by the decreased number of hole carriers (majority carriers) in the valence band during the transport. A large drain voltage bias ($V_d$ in the range of 4 to 5 V) provides enough energy to the electrons trapped at the acceptor states so that they can recombine with holes in the
valence band, which decreases the number of hole carriers and thus the current (Fig. 1G) in the p-type semiconductor (SnS). The responses of the dynamic memristor to electrical pulses are shown in Fig. 1C, where voltage pulses with an amplitude of 4.5 V and a width of 20 ms were used to generate dynamic states in RC. The current level is saturated after 0.5 s, as shown in Fig. 1C, revealing the saturation of the role of acceptor states to the transport. On the other hand, typical photogating occurs in donor states under light illumination. More holes are generated by newly excited electrons and are occupying the donor states as stimulated by light (Fig. 1H).

Dynamic memristor states in RC can be generated by optical pulses, as shown in Fig. 1D. A periodic pulse train with an illumination power of 43 mW and an interval of 3 s (wavelength of 725 nm) produces conductance changes that can be used to realize high-dimensional reservoir states in RC. A read voltage of 0.5 V was used to measure the currents induced by optical stimuli, as shown in Fig. 1D, which is modulated by the photogating effect with a short characteristic time of 3 s. The two different trends shown in Fig. 1 (C, depression, and D, facilitation) reflect the rich charge trapping/detrapping dynamics in 2D SnS with a moderate carrier density of \( \sim 10^{12} \text{ cm}^{-2} \). We note that in other 2D systems apart from certain 2D heterostructures or hybrid materials systems \( (35-45) \), either facilitation or depression dominates the transport, limiting the complexity and multiple-mode operation of optoelectronic RC. Moreover, wide ranges of wavelengths (Fig. 1E and fig. S5) and incident power levels (fig. S6) imply that our optoelectronic RC operates with broadband optoelectronic inputs for various applications.

### Spatiotemporal signal processing in a circuit based on SnS memristors

An integrated memristor circuit based on 2D SnS was fabricated to demonstrate the concept of optoelectronic RC (Fig. 2, A and B). Spatiotemporal optoelectronic inputs are applied to the memristors in the array, as schematically illustrated by the pulses (electrical spikes) and discrete optical beam trains (optical spikes) in Fig. 2A. Such sequential optoelectronic inputs can generate numerous (high-dimensional and dual-mode) reservoir states of RC, which are based on the electrical and optical responses of the dynamic memristors, as shown in Fig. 1. The atomic force microscopy (AFM) image in Fig. 2B shows the topology of a memristor RC.

Modulation of our dynamic memristor, as shown in Fig. 2C, was achieved upon the reception of six input signals: (00100), (01001), (01010), (11101), (01110), and (11111). The response of the device originates from the combined effects of electrical stimulation and fading memory in the SnS-based memristor. The pulse amplitude and width (4 V and 50 ms) representing the input signal “1” were chosen to reflect the characteristic time of the fading memory (100 ms), which helps to generate the distinct states of the memristor shown in Fig. 2C. Starting with a similar current value, the six inputs, each consisting of five electrical pulses, yield six different final current amplitudes. A decrease of the current occurs upon presenting “1” (due to the electrons being detrapped from acceptor states) while the signal “0” tends to recover the conductance (due to the electrons being trapped to acceptor states), as shown in Fig. 1 (F to H).

Therefore, for the electrical inputs, complex memristor states...
(i.e., reservoir states) can be constructed by signals with different combinations of “0” and “1” with a time interval shorter than 100 ms. We note that this is the first demonstration of RC with devices based on 2D materials.

The six optical inputs and corresponding memristor states are presented in Fig. 2D. Following the optical pulses, an electric bias of 0.5 V was used to record the resultant memristor conductance (currents). The six states, corresponding to six different inputs, are clearly distinguishable, which is a critical property to realize optoelectronic RC. We note that similar photoconductance in 2D semiconductors [e.g., molybdenum disulfides (MoS₂)] with high responsivity has also been reported. However, compared to those 2D semiconductors, 2D SnS exhibits better fading memory behavior and is thus able to generate more distinguishable reservoir states based on its opposite and nonlinear responses to electrical and optical stimuli (Fig. 1, C and D, and fig. S4).

Optoelectronic RC for the learning of the Korean language

The integrated memristors were connected to a digital readout layer implemented in software for temporal sequence classification. The sequences consist of the signals representing five complicated consonants and another five vowels of the Korean alphabet, as shown in Fig. 3A. To explain how the system works, a Korean consonant “E” (also similar to the capital English alphabet “E” in appearance) was chosen as an example, as shown in Fig. 3A. The consonant is first divided into five rows, or sequences, as shown in the second panel of Fig. 3A. Then, each sequence is converted to a train of temporal optoelectronic pulses; dark green pixels, which constitute the pattern of the consonant, correspond to the signal “1” (high voltage or optical pulse), while light green pixels of the remaining vacant region of the image only receive read voltages (1 V for 25 ms), as shown in the third panel of Fig. 3A. The signals of the five rows are applied to the five corresponding integrated memristors. The corresponding conductance states (high-dimensional states of the RC system) are schematically illustrated by the orange balls in the fourth panel of Fig. 3A.

The post-training weights for recognizing the 10 consonants and vowels in Fig. 3A are shown in the color maps in Fig. 3B (for electrical stimuli) and Fig. 3C (for optical stimuli). With five memristors (rows) and six temporal signals (sequences including the initial conductance), there are 30 effective reservoir nodes that serve as the inputs to the readout. The colors of the i-th row, consisting of the 31 weights,
including the bias weight, represent the synaptic connections of the ith pattern in the supervised learning. The evolution of the weights is provided in the Supplementary Materials (animated GIF images 1 and 2).

The experimental distribution of individual memristor states (or current levels in our SnS-based memristors) is shown in Fig. 3D. These states are measured after receiving different pulses that are used to represent the 10 input patterns in Fig. 3A. The variation of the final memristor states could be explained by the impact of defects on the transport of SnS, as is widely reported in numerous 2D materials. Optoelectronic RC enables simple and fast training of readout weights at a low computation cost, achieving high accuracy in classifying the 10 temporal inputs (100% in 100 epochs), as shown in Fig. 3E. The losses during the learning of classification of the 10-class temporal inputs with the experimental reservoir states are shown in Fig. S7. The reservoir outputs and associated probability of readout neurons, taking the first letter as an example, are summarized in Fig. S8.

**Inference of Korean sentences via optoelectronic RC**

Optoelectronic RC based on 2D SnS memristors has been further extended to learn Korean sentences, as shown in Fig. 4A: “가자” (Let’s go), “나가” (Get out), “상자” (Let’s buy), “타자” (Let’s ride), and “치다” (Kick). For each sentence, a noise-free image was prepared for training (Fig. 4A), and multiple images with random noises were used for inference (Fig. S9). The image for a sentence is split into five rows. Each of them consists of 13 electric pulses that were sequentially applied to a memristor, which is translated to the high-dimensional states of the optoelectronic RC system. Example responses by the five memristors to electrical inputs are shown in Fig. S10.

The initial and final weights (after 100 training epochs) are shown in Fig. 4 (B and C, respectively). The y axis indicates that the associated five sentences (patterns) and the 71 data points along the x axis (5 × 14 and the bias) correspond to the dimensionality of the reservoir states. The evolution of the weights (readout map) is shown in the Supplementary Materials (animated GIF image 3). As the epoch number during the training process increases, the classification accuracy improves while the cross-entropy loss decreases, as shown correspondingly in Fig. 4 (D and E) with electrical bias as stimuli. The accuracy in classifying test images with random noise (Fig. S9) is as high as 91% for the practical Korean sentences used here. The accuracy is smaller (91%) in Fig. 4D because the patterns (the short Korean sentences) in Fig. 4D are much more complex compared with those used for classification in Fig. 3E. Moreover, for the short Korean sentences of Fig. 4A, intentional background noise (shown in Fig. S9) was introduced, which corresponds to the classification accuracy in Fig. 4D. The inference samples in Fig. S9 were chosen to be different from those used to train the network.
As a proof of concept, we achieved an accuracy of 91% during the classification of five practical Korean sentences each containing intentional noise.

Our finding overcomes the hardware bottleneck associated with physically separated sensors and processors, where analog-to-digital data conversion incurs large energy consumption and latency penalties in conventional sensing–computing systems. In addition, RC in this case greatly reduces the learning complexity. This energy-efficient method enables efficient machine learning applications with temporal inputs at the edge, an advance in strong demand in the era of the IoT. Compared with biological neurons of different/specialized sensing modes, our dual-mode processing could be considered as a breakthrough for efficient machine learning and neuromorphic computing.

**DISCUSSION**

The unique electrical and optical responses of 2D SnS, originating from its rich defect states with both Sn and S atomic vacancies, have been successfully adopted for the novel in-sensor RC for language learning with dual-mode operation. The unique and opposite trends of the electrical and optical responses of 2D SnS enable robust learning of the Korean language.

As shown in Fig. 4A; thus, the accuracy is lowered in Fig. 4D compared with that shown in Fig. 3E.

A confusion matrix showing the classification of the five sentences is presented in Fig. 4F. Nearly all of the sentences were correctly classified by our optoelectronic RC method. We note that the first and third sentences exhibit weak confusion, but recognition overall is not disturbed by the slightly deviating performance. As an example, the outputs of reservoir neurons, or the reservoir states, for the second sentence “$\text{ㄱ}{\text{ㅏ}}$” and the probability of the readout neurons (indexed following the five sentences in Fig. 4A) after the training are shown in Fig. 4 (G and H, respectively).

The robustness of our optoelectronic RC approach is demonstrated in Fig. 5. To account for the stochasticity of the neurons, we model the neuron output currents with Gaussian distributions of varying standard deviations (SDs) ($\sigma$ ranging from 0.1 to 0.6 nA) (Fig. 5A and fig. S11) and simulated the classification accuracy. The first sentence in Fig. 4A, “$\text{ㄱ}{\text{ㅏ}}$,” is taken as an example in Fig. 5A. Three commonly used classification methods that can implement the readout functions were compared. Each method is applied to the six sets of RC neural outputs with different standard deviations shown in Fig. 5B. Language learning via the proposed optoelectronic RC method exhibits high accuracy rates even with the noisy reservoir node outputs; the accuracy levels of all classification methods exceed 85% with SDs up to $\sigma = 0.6$ nA for the different models. Accordingly, the high-dimensional states and the nonlinear evolution of our optoelectronic RC method based on 2D SnS enable robust learning of the Korean language.

**MATERIALS AND METHODS**

**Device fabrication details**

SnS flakes were exfoliated from the original crystal and transferred onto a silicon wafer substrate with 300 nm of SiO$_2$ covered by dry oxidation method. Spin coating with poly(methylmethacrylate) 950 A4 was then carried out at a spin speed of 500 rpm for the first 5 s and of 3000 rpm for the following 60 s. The wafer was subsequently baked at 120°C on a commercial hot plate for 2 min. Exposure of the electrode pattern was carried out by electron beam lithography followed by a standard developing process. Last, Cr/Au 5/100-nm deposition was done using an electron beam evaporator.

**Electrical/optical characteristics**

Electrical characterization was conducted with a Keithley 4200 semiconductor parameter analyzer and a Cascade probe station. For single-device measurements, the pulse condition is 4.5 V/20 ms. The read voltage is 1 V. The training of reservoir states is based on 11 different inputs relative to the five consonants and five vowels used in this work. For the training of the memristor array, the pulse condition is 4 V/50 ms and the read pulse condition is 1 V/25 ms. Four laser line wavelengths (455, 638, 725, and 811 nm) were used for testing.
Simulations
Softmax regression was used to fit the weights of the readout layer. The categorical cross-entropy loss is minimized by batch gradient descent using the RMSprop (learning ratio = 0.01) optimizer. The accuracies of the SD-dependent feature classification shown in Fig. 5B were obtained as follows. The amounts of the current flowing through five memristors in the reservoir at the final time step were used as the inputs to the readout layer. Supervised learning models like single-layer perceptron, support vector machine, and logistic regression were used for the learning of the layer. For the single-layer perceptron, Softmax was used as the activation function. The categorical cross-entropy loss was minimized by a mini-batch (the batch size is 16) gradient descent using the Adam optimizer (learning rate = 0.001). For the support vector machine, the kernel option was chosen to be linear for feature classification. Regularization parameter $c$ was used, whose value was optimized by grid search. For the logistic regression, regularization parameters elasticnet and $c$ were used with grid search for fine-tuning.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/20/eabg1455/DC1

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Sun et al., Sci. Adv. 2021; 7 : eabg1455 14 May 2021 7 of 8
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