

Article

Spike-Timing-Dependent Plasticity Device with Ga-Sn-O Conductance Change Layer Deposited by Mist-CVD Method

Hidehito Kita ¹, Kazuma Uno ², Tokiyoshi Matsuda ^{3,4}, Hidenori Kawanishi ^{4,5} and Mutsumi Kimura ^{1,2,5,*} 

¹ Graduate School of Advanced Science and Technology, Ryukoku University, Otsu 520-2194, Seta, Japan; y24m026@mail.ryukoku.ac.jp

² Faculty of Advanced Science and Technology, Ryukoku University, Otsu 520-2194, Seta, Japan; ryukokuronbun@gmail.com

³ Graduate School of Science and Engineering, Kindai University, Higashiosaka 577-8502, Osaka, Japan; matsuda@ele.kindai.ac.jp

⁴ Innovative Materials and Processing Research Center, Ryukoku University, Otsu 520-2194, Seta, Japan; temp220102@mail.ryukoku.ac.jp

⁵ Graduate School of Science and Technology, Nara Institute of Science and Technology (NAIST), Ikoma 630-0192, Nara, Japan

* Correspondence: mutsu@rins.ryukoku.ac.jp

Abstract: A spike-timing-dependent plasticity (STDP) device with a Ga-Sn-O (GTO) conductance change layer deposited by a mist-CVD method has been developed. First, the memristive characteristic is analyzed. Next, based on it, spike waveforms are determined. Finally, the STDP characteristic is successfully confirmed. This is an original report on the realization of an STDP characteristic using a thin film deposited by the mist-CVD method, which is achieved by the GTO properties and a well-designed clear methodology to realize a STDP characteristic from a memristive characteristic.

Keywords: spike-timing-dependent-plasticity (STDP); Ga-Sn-O (GTO); mist-CVD; memristive characteristic



Citation: Kita, H.; Uno, K.; Matsuda, T.; Kawanishi, H.; Kimura, M.

Spike-Timing-Dependent Plasticity Device with Ga-Sn-O Conductance Change Layer Deposited by Mist-CVD Method. *Electronics* **2024**, *13*, 3413. <https://doi.org/10.3390/electronics13173413>

Academic Editor: Ray-Hua Horng

Received: 14 July 2024

Revised: 6 August 2024

Accepted: 27 August 2024

Published: 28 August 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Artificial intelligence (AI) provides critical infrastructures in modern and future societies through diverse applications [1,2]. Neural networks are the most popular embodiments of AI that mimic the operating theories of living brains [3,4]. However, traditional models of neural networks are large and complex software that runs on high-spec and energy-consuming hardware such as Neumann-type digital computers, which is not customized for neural networks [5,6]. Neuromorphic systems are practical solutions that consist only of customized devices, circuits, and hardware for neural networks with compact computer size and lower energy consumption [7,8]. Memristors are emerging devices that can be used as not only digital memories in old-fashioned computers [9,10] but also analog memories in neuromorphic systems [11–20]. However, once memristors are integrated into neuromorphic systems as computing in memory (CIM) or processing in memory (PIM) [21], it is considerably difficult to integrate control circuits to adjust electrical conductivities because they are likely more complicated than neuromorphic systems themselves to generate arbitrary voltage or current. Therefore, autonomous learning is strongly desired. Incidentally, spiking neuromorphic systems are notable schemes as extremely low-energy systems, where spike pulses are utilized to send signals [22]. Spike-timing-dependent plasticity (STDP) devices are inspired by a biomimetic approach from living brains and promise to enable autonomous learning in spiking neuromorphic systems, and STDP characteristics are often realized as more advanced utilizations of memristor characteristics [23–27]. However, it was sometimes difficult to realize them, because there are many candidates of component materials, device structures, fabrication processes, driving schemes, etc.

Semiconductor materials are key points for actual fabrication of memristors. Amorphous metal-oxide semiconductors (AOSs) are semiconductor materials using common and inexpensive chemical elements and can exhibit excellent characteristics even if they are deposited at low temperatures, and therefore they are superior in low material and fabrication cost [28]. In addition, deposition methods are other key points for actual fabrication of memristors. Mist chemical vapor deposition (CVD) methods are deposition methods in atmosphere pressure, and they are also superior in terms of the low fabrication cost [29,30]. However, it was not reported that an STDP device can be realized with an AOS thin film deposited by the mist CVD method.

In this research, an STDP device with a Ga-Sn-O (GTO) conductance change layer deposited by a mist CVD method has been developed. GTO is an AOS and is superior in terms of the low material cost, non-toxicity, etc. [31,32]. The device details, fabrication processes, memristive characteristics, spike waveforms, and STDP characteristic will be explained. This is an original report on the well-designed combination of the STDP, GTO, and mist CVD technologies, and provides a clear methodology to realize an STDP characteristic from a memristive characteristic. In summary, this research has achieved two specific goals. The first goal is to develop an STDP device with low material and fabrication costs without noticeable improvement in the memristive characteristic, even if it is poor. The second goal is to provide a methodology to realize an STDP characteristic, again, even if the memristive characteristic is poor, and no previous work has provided such a clear methodology.

2. STDP Device with a GTO Conductance Change Layer Deposited by a Mist CVD Method

An STDP device with a GTO conductance change layer deposited by a mist CVD method is shown in Figure 1. First, the device structure and fabrication processes are shown in Figure 1a. A quartz substrate of the size of 15 mm × 15 mm and thickness of 0.7 mm is used, and an Al thin film is deposited using a vacuum evaporation method through a metal mask as a bottom electrode. Next, the hot-wall-type mist CVD method is shown in Figure 1b. The GTO thin film is deposited using the mist CVD method as a conductance change layer. Ga acetylacetonate (Ga(acac)) of 0.0855 g and Sn acetylacetonate (Sn(acac)) of 0.3324 g are dissolved in HCl of 20% and 3 g and pure water of 20 mL, whose volumes are adjusted so that the atomic composition is Ga:Sn = 1:4.5 in the solution. A mist of the solution is generated by ultrasonic generators, carried by a carrier gas of air of a flow rate of 1 L/min, diluted by a dilution gas of air of a flow rate of 1 L/min, and injected through a quartz tube, and the inside of the quartz tube is heated to the temperature of 450 °C by a heater. The GTO thin film is deposited during the deposition time of 30 min to the film thickness of 33 nm, which is measured using a stylus profiler. It is believed that the oxidizer is acac. The atomic composition may deviate from the stoichiometry, and defect states such as oxygen vacancies are formed and become the source elements of electrical conduction. Finally, a Au thin film is deposited using a vacuum evaporation method through a metal mask as a top electrode. The device structure is quite simple, namely, the GTO thin film is just sandwiched between the top and bottom electrodes.

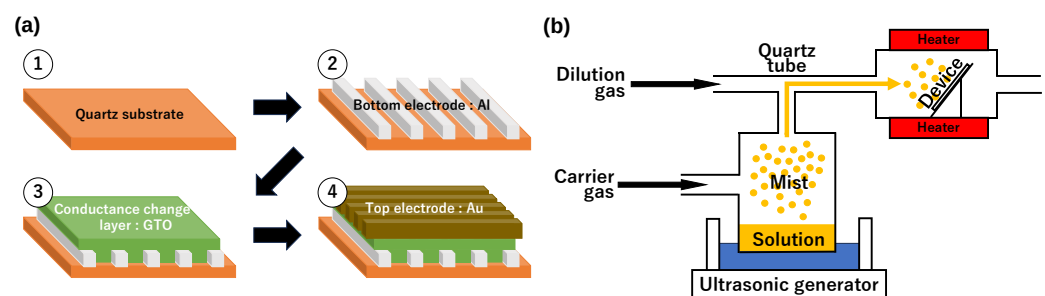


Figure 1. STDP device with a GTO conductance change layer deposited by a mist CVD method. (a) Device structure and fabrication processes. (b) Hot-wall-type mist CVD method.

3. Memristive Characteristic

The memristive characteristic is shown in Figure 2. First, the hysteresis characteristic is shown in Figure 2a. Here, the bottom electrode is grounded, and a voltage applied to the top electrode (V) is scanned to 0~−4.5 V~0 as a reset operation. Then, V is scanned to 0~+V_{set}~0 with varying V_{set} = 2~4 V as set operations, and the flowing current (I) is measured. The magnified graph is also superimposed when V is 0~1 V. It is found that the set operation induces the transition from a low conductance state (LCS) to a high conductance state (HCS), whereas the reset operation initializes the electrical conductance. Hysteresis curves are observed, and the width of the hysteresis increases as V_{set} increases. Next, the switching ratio is shown in Figure 2b. Here, the switching ratio is defined as the conductance ratio between the HCS and LCS for V = 0.5 V. It is analyzed that the switching ratio remains the same below V_{set} ≤ 3 V and increases as V_{set} increases above V_{set} ≤ 3.5 V. The endurance and retention characteristics are under evaluation and expected to be not so bad based on previous results [33], so we would like to report them in the near future. The switching mechanism is also shown in [33], which is the reason why Al thin film is used as a bottom electrode, whereas Au thin film is used as a top electrode.

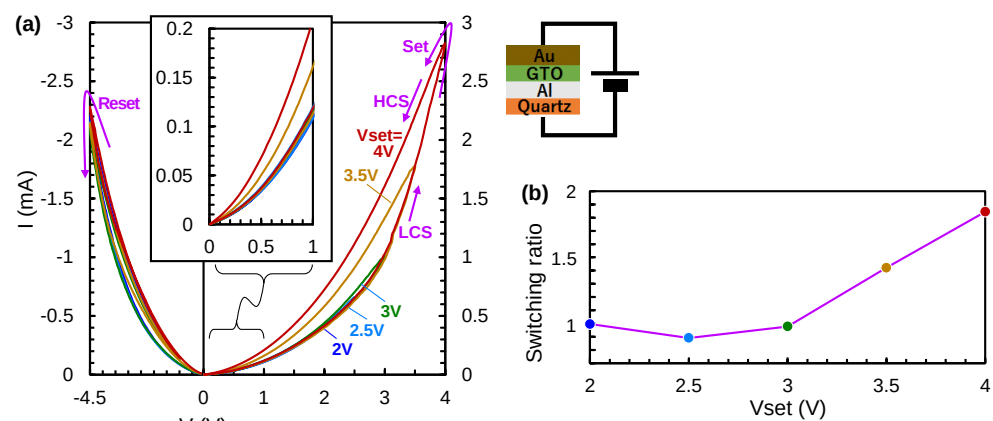


Figure 2. Memristive characteristic. (a) Hysteresis characteristic. (b) Switching ratio.

4. Spike Waveforms

Based on the memristive characteristic shown in Figure 2b, spike waveforms are determined. The spike waveforms are shown in Figure 3a. First, pre-spike voltage (V_{pre}), post-spike voltage (V_{post}), and net voltage (V_{net}) are shown in Figure 3a. Here, V_{pre} is the voltage that is supposed to be applied to the top electrode, V_{post} is the voltage applied to the bottom electrode, and V_{net} = V_{pre} − V_{post} is the voltage applied to the STDP device. Δt is defined as the time lag between the V_{pre} and V_{post}, and the spike waveforms for Δt = −0.2, 0, and +0.2 ms are shown as examples. Next, the maximum voltage of the V_{net} (V_{max}) is shown in Figure 3b. It is found that the V_{max} ranges from V_{max} ≤ 3 V to V_{max} ≥ 3.5 V. Conversely, the spike waveforms are well designed so that the V_{max} is so. As a result, it is expected that the device conductance will remain the same or increase depending on the Δt.

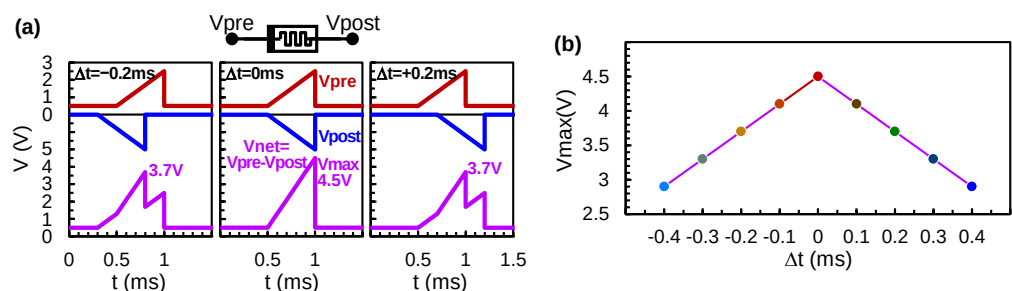


Figure 3. Spike waveforms. (a) V_{pre}, V_{post}, and V_{net}. (b) V_{max}.

5. STDP Characteristic

The STDP characteristic is shown in Figure 4. The conductance change along time (t) is shown in Figure 4a. Here, the V_{pre} and V_{post} shown in Figure 3a are actually applied to the top and bottom electrodes, respectively, with a frequency of 2 kHz, and the device conductance is measured in situ because small bias is applied when no spike pulse is applied, as shown in Figure 3a. The conductance change is calculated as the change from the initial value. It is found that the device conductance almost remains the same for $\Delta t = \pm 0.4$ ms, increases for $\Delta t = \pm 0.2$ ms, and further increases for $\Delta t = 0$ ms, which is as expected. Next, the conductance change and switching ratio are shown in Figure 4b. Here, the conductance change is obtained from that for $t = 9$ min in Figure 4a because it seems to roughly saturate, and the switching ratio is calculated by relating the V_{set} shown in Figure 2b and V_{max} shown in Figure 3b. For example, $V_{max} = 3.7$ V for $\Delta t = 0.2$ ms in Figure 3b, and the switching ratio is read as 1.59 for this V_{max} in Figure 2b. It is found that the tendency of the conductance change is quite similar to that of the switching ratio, which is as expected. As a result, the STDP characteristic is successfully confirmed.

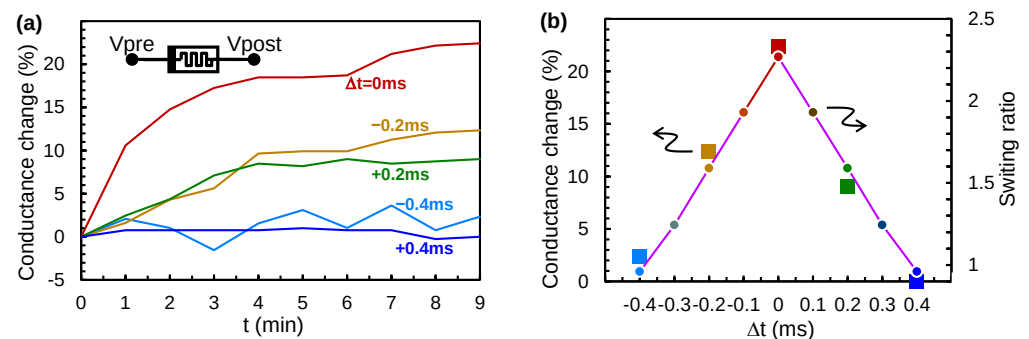


Figure 4. STDP characteristic. (a) Conductance change along t . (b) Conductance change and switching ratio.

6. Conclusions

An STDP device with a GTO conductance change layer deposited by a mist CVD method has been developed. First, the memristive characteristic was analyzed. Next, based on it, spike waveforms were determined. Finally, the STDP characteristic was successfully confirmed. This is an original report on the realization of an STDP characteristic using a thin film deposited by the mist CVD method, which is achieved by the GTO properties and a well-designed clear methodology to realize an STDP characteristic from a memristive characteristic. As mentioned above, this research has achieved two specific goals. The first goal was to develop an STDP device with low material and fabrication costs without noticeable improvement in the memristive characteristic, even if it is poor. The second goal was to provide a methodology to realize an STDP characteristic, again even if the memristive characteristic is poor. In other words, how to realize the STDP characteristic from the memristive characteristic was proposed, which will be useful for all future researchers who would like to apply their memristive devices to STDP devices. We expect that not only STDP devices we develop but also those that other researchers develop can find possible applications for neuromorphic systems.

Author Contributions: Conceptualization, H.K. (Hidehito Kita) and M.K.; methodology, H.K. (Hidehito Kita) and K.U.; software, H.K. (Hidehito Kita); validation, H.K. (Hidenori Kawanishi) and M.K.; formal analysis, T.M.; investigation, H.K. (Hidehito Kita) and M.K.; resources, H.K. (Hidehito Kita) and K.U.; data curation, H.K. (Hidehito Kita) and K.U.; writing—original draft preparation, H.K. (Hidehito Kita) and M.K.; writing—review and editing, H.K. (Hidehito Kita) and M.K.; visualization, H.K. (Hidehito Kita) and M.K.; supervision, H.K. (Hidenori Kawanishi) and M.K.; project administration, M.K.; funding acquisition, M.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by JSPS KAKENHI (A) 22H00515, (C) 19K11876, JST Taiwan, ALCA-Next, High-Tech Research Center in Ryukoku University, Laboratory for Materials and Structures in Tokyo Institute of Technology, and Research Institute of Electrical Communication in Tohoku University.

Data Availability Statement: Data is contained within the article.

Conflicts of Interest: The authors declare no conflicts of interest.

References

- McCarthy, J.; Minsky, M.L.; Rochester, N.; Shannon, C.E. A proposal for the Dartmouth Summer Research Project on artificial intelligence. *AI Mag.* **1955**, *27*, 12.
- Zhang, C.; Lu, Y. Study on artificial intelligence: The state of the art and future prospects. *J. Ind. Inf. Integr.* **2021**, *23*, 100224. [\[CrossRef\]](#)
- McCulloch, W.S.; Pitts, W. A logical calculus of the ideas immanent in nervous activity. *Bull. Math. Biophys.* **1943**, *5*, 115–133. [\[CrossRef\]](#)
- Aggarwal, C.C. *Neural Networks and Deep Learning: A Textbook*; Springer: Berlin/Heidelberg, Germany, 2018.
- Greenemeier, L. Will IBM's Watson Usher in a New Era of Cognitive Computing? *Scientific American*, 13 November 2013.
- Lohr, S. What ever happened to IBM's Watson? *The New York Times*, 16 July 2021.
- Lande, T.S. *Neuromorphic Systems Engineering, Neural Networks in Silicon*; Springer Science & Business Media: Berlin, Germany, 2013.
- Mohamed, K.S. *Neuromorphic Computing and Beyond: Parallel, Approximation, Near Memory, and Quantum*; Springer: Cham, Switzerland, 2020.
- Strukov, D.B.; Snider, G.S.; Stewart, D.R.; Williams, R.S. The missing memristor found. *Nature* **2008**, *453*, 80–83. [\[CrossRef\]](#) [\[PubMed\]](#)
- Chen, Y. ReRAM: History, status, and future. *IEEE Trans. Electron Devices* **2020**, *67*, 1420–1433. [\[CrossRef\]](#)
- Prezioso, M.; Merrih-Bayat, F.; Hoskins, B.D.; Adam, G.C.; Likharev, K.K.; Strukov, D.B. Training and operation of an integrated neuromorphic network based on metal-oxide memristors. *Nature* **2015**, *521*, 61–64. [\[CrossRef\]](#)
- Yao, P.; Wu, H.; Gao, B.; Tang, J.; Zhang, Q.; Zhang, W.; Yang, J.J.; Qian, H. Fully hardware-implemented memristor convolutional neural network. *Nature* **2020**, *577*, 641–646. [\[CrossRef\]](#)
- Simanjuntak, F.M.; Ohno, T.; Chandrasekaran, S.; Tseng, T.-Y.; Samukawa, S. Neutral oxygen irradiation enhanced forming-less ZnO-based transparent analog memristor devices for neuromorphic computing applications. *Nanotechnology* **2020**, *31*, 26LT01. [\[CrossRef\]](#)
- Zeng, F.; Guo, Y.; Hu, W.; Tan, Y.; Zhang, X.; Feng, J.; Tang, X. Opportunity of the lead-free all-inorganic Cs₃Cu₂I₅ perovskite film for memristor and neuromorphic computing applications. *ACS Appl. Mater. Interfaces* **2020**, *12*, 23094–23101. [\[CrossRef\]](#)
- Das, U.; Sarkar, P.; Paul, B.; Roy, A. Halide perovskite two-terminal analog memristor capable of photo-activated synaptic weight modulation for neuromorphic computing. *Appl. Phys. Lett.* **2021**, *118*, 182103. [\[CrossRef\]](#)
- Hu, L.; Yang, J.; Wang, J.; Cheng, P.; Chua, L.O.; Zhuge, F. All-optically controlled memristor for optoelectronic neuromorphic computing. *Adv. Funct. Mater.* **2021**, *31*, 2005582. [\[CrossRef\]](#)
- Peng, Z.; Wu, F.; Jiang, L.; Cao, G.; Jiang, B.; Cheng, G.; Ke, S.; Chang, K.-C.; Li, L.; Ye, C. HfO₂-based memristor as an artificial synapse for neuromorphic computing with tri-layer HfO₂/BiFeO₃/HfO₂ design. *Adv. Funct. Mater.* **2021**, *31*, 2107131. [\[CrossRef\]](#)
- Li, Y.; Ang, K.-W. Hardware implementation of neuromorphic computing using large-scale memristor crossbar arrays. *Adv. Intell. Syst.* **2021**, *3*, 2000137. [\[CrossRef\]](#)
- Meng, J.-L.; Wang, T.-Y.; He, Z.-Y.; Chen, L.; Zhu, H.; Ji, L.; Sun, Q.-Q.; Ding, S.-J.; Bao, W.-Z.; Zhou, P.; et al. Flexible boron nitride-based memristor for in situ digital and analogue neuromorphic computing applications. *Mater. Horiz.* **2021**, *8*, 538–546. [\[CrossRef\]](#) [\[PubMed\]](#)
- Park, S.-O.; Jeong, H.; Park, J.; Bae, J.; Choi, S. Experimental demonstration of highly reliable dynamic memristor for artificial neuron and neuromorphic computing. *Nat. Commun.* **2022**, *13*, 2888. [\[CrossRef\]](#)
- Milojicic, D.; Bresnaker, K.; Campbell, G.; Faraboschi, P.; Strachan, J.P.; Williams, S. Computing In-Memory, Revisited. In Proceedings of the 2018 IEEE 38th International Conference on Distributed Computing Systems (ICDCS), Vienna, Austria, 2–6 July 2018; pp. 1300–1309. [\[CrossRef\]](#)
- Maass, W. Networks of spiking neurons: The third generation of neural network models. *Neural Netw.* **1997**, *10*, 1659–1671. [\[CrossRef\]](#)
- Bi, G.Q.; Poo, M.M. Synaptic modifications in cultured hippocampal neurons: Dependence on spike timing, synaptic strength, and postsynaptic cell type. *J. Neurosci.* **1998**, *18*, 10464–10472. [\[CrossRef\]](#)
- Wittenberg, G.M.; Wang, S.S.H. Malleability of spike-timing-dependent plasticity at the CA3–CA1 synapse. *J. Neurosci.* **2006**, *26*, 6610–6617. [\[CrossRef\]](#)
- Buchanan, K.A.; Mellor, J.R. The development of synaptic plasticity induction rules and the requirement for postsynaptic spikes in rat hippocampal CA1 pyramidal neurons. *J. Physiol.* **2007**, *585*, 429–445. [\[CrossRef\]](#)
- Buchanan, K.A.; Mellor, J.R. The activity requirements for spike timing-dependent plasticity in the hippocampus. *Front. Synaptic Neurosci.* **2010**, *2*, 11. [\[CrossRef\]](#)

27. Serrano-Gotarredona, T.; Masquelier, T.; Prodromakis, T.; Indiveri, G.; Linares-Barranco, B. STDP and STDP variations with memristors for spiking neuromorphic learning systems. *Front. Neurosci.* **2013**, *7*, 2. [[CrossRef](#)] [[PubMed](#)]
28. Nomura, K.; Ohta, H.; Takagi, A.; Kamiya, T.; Hirano, M.; Hosono, H. Room-temperature fabrication of transparent flexible thin-film transistors using amorphous oxide semiconductors. *Nature* **2004**, *432*, 488–492. [[CrossRef](#)] [[PubMed](#)]
29. Lu, J.-G.; Kawaharamura, T.; Nishinaka, H.; Kamada, Y.; Oshima, T.; Fujita, S. Zn-based thin films synthesized by atmospheric pressure mist chemical vapor deposition. *J. Cryst. Growth* **2007**, *299*, 1–10. [[CrossRef](#)]
30. Dang, G.T.; Kawaharamura, T.; Furuta, M.; Allen, M.W. Metal-semiconductor field-effect transistors with In–Ga–Zn–O channel grown by nonvacuum-processed mist chemical vapor deposition. *IEEE Electron Device Lett.* **2015**, *36*, 463–465. [[CrossRef](#)]
31. Matsuda, T.; Umeda, K.; Kato, Y.; Nishimoto, D.; Furuta, M.; Kimura, M. Rare-metal-free high-performance Ga–Sn–O thin film transistor. *Sci. Rep.* **2017**, *7*, 44326. [[CrossRef](#)]
32. Sugisaki, S.; Matsuda, T.; Uenuma, M.; Nabatame, T.; Nakashima, Y.; Imai, T.; Magari, Y.; Koretomo, D.; Furuta, M.; Kimura, M. Memristive characteristic of an amorphous Ga–Sn–O thin-film device. *Sci. Rep.* **2019**, *9*, 2757. [[CrossRef](#)]
33. Takishita, Y.; Kobayashi, M.; Hattori, K.; Matsuda, T.; Sugisaki, S.; Nakashima, Y.; Kimura, M. Memristor property of an amorphous Sn–Ga–O thin-film device deposited using mist chemical-vapor-deposition method. *AIP Adv.* **2020**, *10*, 035112. [[CrossRef](#)]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.