

2D MoS 2 Monolayers Integration with Metal Oxide-based Artificial Synapses

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Scope Statement

The presented work is physical modeling of the memristive device based on the metal oxide with its integration with 2D monolayers MOS2 to establish the artificial synapses application with low power operations. This work enables the future scope of metal oxide memristive devices and their practical integration with 2D materials to design and fabricate new types of memories for various applications.

Conflict of interest statement

The authors declare a potential conflict of interest and state it below

The author(s) declared that they were an editorial board member of Frontiers, at the time of submission. This had no impact on the peer review process and the final decision

CRediT Author Statement

Dimitra G. Georgiadou: Formal Analysis, Funding acquisition, Investigation, Project administration, Resources, Supervision, Validation, Visualization, Writing - review & editing. Ioannis Zeimpekis: Funding acquisition, Investigation, Project administration, Resources, Software, Supervision, Visualization, Writing - review & editing. Mohit Kumar: Conceptualization, Data curation, Formal Analysis, Methodology, Software, Validation, Writing - review & editing. Dr. Sanjay Kumar: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing. Shalu Rani: Formal Analysis, Investigation, Visualization, Writing - review & editing.

Keywords

2D-TMD materials, Metal oxide materials, Layered integration, Memristive Devices, Artificial synapses

Abstract

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In this study, we report on a memristive device structure wherein monolayers of two-dimensional (2D) molybdenum disulfide (MoS2) are integrated with an ultrathin yttrium oxide (Y2O3) layer to simulate artificial synapses functionality. The proposed physical simulation methodology is implemented in COMSOL Multiphysics tool and is based on the minimization of free energy of the used materials at the applied input voltage. The simulated device exhibits a stable bipolar resistive switching (RS) and the switching voltages is significantly reduced by increasing the number of MoS2 layers, which is key to conventional low-power computing and neuromorphic applications. The device is shown to perform synaptic functionalities under various applied bias conditions. The resulting synaptic weight decreases almost linearly with the increasing number of MoS2 layers due to the increase in the device thickness. The simulation outcomes pave the way for the development of optimised metal oxide-based memristive devices through their integration with semiconducting 2D materials. Also, the 2D MoS2 integration can enable the optoelectronic operation of this memory device.

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Studies involving animal subjects

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2D MoS₂ Monolayers Integration with Metal Oxide-based Artificial Synapses

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Abstract: In this study, we report on a memristive device structure wherein monolayers of two-dimensional (2D) molybdenum disulfide (MoS2) are integrated with an ultrathin yttrium oxide (Y2O3) layer to simulate artificial synapses functionality. The proposed physical simulation methodology is implemented in COMSOL Multiphysics tool and is based on the minimization of free energy of the used materials at the applied input voltage. The simulated device exhibits a stable bipolar resistive switching (RS) and the switching voltages is significantly reduced by increasing the number of MoS2 layers, which is key to conventional low-power computing and neuromorphic applications. The device is shown to perform synaptic functionalities under various applied bias conditions. The resulting synaptic weight decreases almost linearly with the increasing number of MoS2 layers due to the increase in the device thickness. The simulation outcomes pave the way for the development of optimised metal oxide-based memristive devices through their integration with semiconducting 2D materials. Also, the 2D MoS2 integration can enable the optoelectronic operation of this memory device.

Index Terms: 2D-TMD materials, Metal Oxide materials, Layered integration, Memristive devices, Artificial synapses.

1. INTRODUCTION

Emerging non-volatile memories such as memristive devices that can act as artificial synapses, have attracted huge interest recently in the field of neuromorphic computing, due to their unique capabilities, including high-density integration, fast write and read speed, and compatibility with the existing complementary metal oxide semiconductor process [1-4]. In memristive devices data can be written, processed, and erased by changing resistance states between one or multiple low resistance (LRS) and high resistance states (HRS), while the original resistance state, usually, remains unchanged [5-7].

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Several material systems, including transition metal oxides (TMOs), such as HfO₂ [8], Y₂O₃ [9-14], Ta₂O₅ [15], and TiO₂ [16]), and 2D transition metal dichalcogenides (TMDs), like MoS₂ [17], WS₂ [18], and MoSe₂ [19], have been extensively employed to develop memristive devices for numerous applications [17-19]. Notably, 2D TMD materials offer outstanding electronic, optical, and mechanical properties [20-21], as compared to conventional TMO materials, which further enables significant advantages [22-24], including low-power switching [25-29], device thermal stability [30], electrostatic tunability [31], and mechanical flexibility [20]. Therefore, the integration of layered 2D TMD materials with TMO materials in memristive devices can further enhance thermal stability, induce switching at lower voltages, and enable low power/energy operation [32-35]. Moreover, such memristive structures can be utilized to emulate artificial synapses by mimicking basic synaptic functions, such as potentiation, depression and voltage-dependent synaptic responses.

Several successful attempts have been demonstrated to physically model the metal oxide-based memristive devices. For example, Kim et al [36] have reported a physical electro-thermal model of Ta₂O₅ bilayer-based resistive memory by utilizing COMSOL Multiphysics. Bocquet et al [37] and Traore et al [38] have presented the physics-based compact numerical models and density function theory (DFT)-based calculation by adopting generalized gradient approximation (GGA) and Perdew-Burke-Ernzerhof functional (PBE) for HfO2-based RRAM [38], respectively. Kumar et al [39] have implemented an Y2O3-based nanoscale memristor emulating synaptic functionality. However, aforementioned metal oxide-only based memristive devices have several disadvantages, including thermal instability [40], high switching voltage [41], and limited tunability of the device conductance.

In this work, we propose an integration of a number of 2D MoS_2 layers with ultrathin Y_2O_3 in a memristive device structure that is operated as artificial synapse. Here, it should be noted that the integration of 2D MoS_2 with metal oxide memristive structure can be implemented in optoelectronic memories and may also act as optoelectronic synapse for bionic visual applications. The proposed physical model is based on the quantitative thermodynamic numerical modelling of the memristive devices, while COMSOL Multiphysics is used to model the memristive device operation. COMSOL Multiphysics [42] helps to solve the partial differential equations by utilizing Finite Element Method (FEM). The heat transfer and electrodynamic equations

have been resolved by utilizing physical parameters of $2D \text{ MoS}_2$ and Y_2O_3 .

2. MODELLING WORK

2.1 Memristive Device Structure and Adopted Numerical Methodology

In this physical modelling process, an Al (top electrode, TE, 30 nm)/MoS₂ (1-3 layers)/Y₂O₃ (5 nm)/Al (bottom electrode, BE, 50 nm) memristive structure is analysed having a cross-sectional area of 314 nm². Fig. 1(a-c) shows the simulated device structure wherein the 2D MoS₂ layers are varied from 1 to 3 with corresponding film thickness. As can be seen in Fig. 1(a-c), a layer of SiO₂ (with a width of 500 nm) is also utilised as a heat shield layer surrounding the memristive device. The thickness of Al top and bottom electrodes is 30 and 50 nm, respectively, while a thin Y₂O₃ (5 nm) layer is used as the primary resistive switching (RS) layer. We introduce in this structure the 2D MoS₂ monolayer(s), which are known to offer higher thermal conductivity, as compared to transition metal oxide materials [43-46]. Therefore, this structure is expected to contribute to improved thermal stability of the device and to enable low-power in-memory computation by reducing the device switching voltage. Furthermore, this device structure also enables the concept of the optoelectronic memories (as 2D materials have more attractive optical properties, as compared to metal oxides) that can be used in various applications, including optical in-memory computing sensors, and bionic visual systems.

The thermodynamic numerical analysis model that was used relies on the principle of minimizing free energy (FE) within a memristive device, as FE varies under the

influence of an external voltage. Simultaneously, the device reduces its free energy by utilizing phase transitions in the oxide material, involving the breaking of chemical bonds. This free energy within a memristive device is expressed as follows [47]:

$$F = \int \rho C_{P} \, \delta T dx^{3} + \frac{1}{2} \int \varepsilon |E|^{2} dx^{3} + 2\pi r h \sigma_{S} + \pi r^{2} h \delta \mu \tag{1}$$

Herein, ρ : mass density of materials used in memristive structure, $C_{\rm P}$: specific heat capacity of materials used in memristive structure at constant pressure, δT : change in temperature due to variations in thermal gradient inside the device, ε : permittivity of the active materials utilized in the memristive device, E: electric field, r: radius of conductive filament (CF), h: CF height in SET process, l: gap length in the case of RESET process, σ_s : interfacial energy, and $\delta\mu$: difference in chemical potential between unstable conductive phase and insulating phase in SET process ($\delta\mu_1$) and between unstable conductive phase and metastable conductive phase in RESET process ($\delta\mu_2$) [47-48].

Equation (1) delineates thermal and electrostatic energies in its first and second terms, respectively. The latter two terms correspond to phase transformation energy. Notably, the electrostatic energy stemming from the conductive elements (electrodes and filament) is marginal compared to the insulating layer, which possesses higher capacitance. Consequently, the insulator layer predominantly influences the overall free energy. Conversely, the thermal contribution is primarily driven by the conducting filament facilitating current flow between the top and bottom electrodes.

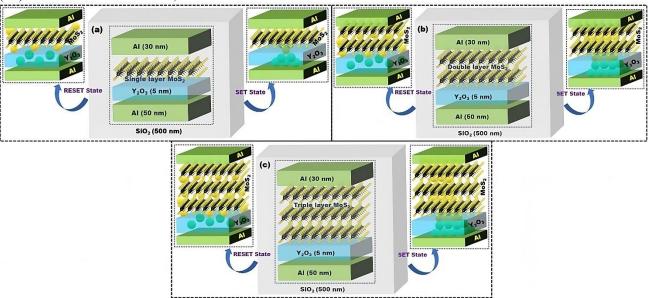


Fig. 1: 3D structure of 2D MoS_2 integrated thin Y_2O_3 -based memristive device integrated with (a) single layer MoS_2 , (b) double layer MoS_2 , and (c) triple layer MoS_2 . Insets of each section show the SET and RESET operations wherein the dominant resistive switching mechanism is the formation and rupture of the CFs.

This work utilizes the following algorithmic steps to determine the device's minimum free energy configuration and its corresponding current-voltage (I-V) characteristics: (a) Construct the device. (b) Apply a

source voltage and compute the device's free energy for varying filament radii. (c) Determine the change in free energy $(\partial F/\partial r)$ corresponding to filament radius and gap length $(\partial F/\partial l)$ for a constant source voltage. (d) Repeat

steps (b) and (c) for different source voltages. (e) Record the device voltage, current, filament radius, filament gap length, and their respective minimum free energies for all source voltages. (f) Finally, obtain two sets of current-voltage (*I-V*) characteristics in step (e) pertaining to the SET and RESET processes.

The SET process comprises two key sub-processes: the rapid shunting of electrodes and the radial expansion of conductive filaments (CFs). Within the shunting phase, there are distinct stages: nucleation and longitudinal growth of CFs, both characterized by their stochastic behaviour [49]. Likewise, the RESET process consists of two fundamental sub-processes: the initiation of a gap through CF rupturing, followed by the stochastic growth of this gap.

2.2. COMSOL Multiphysics Modules

Fig. 1(a-c) depicts the 3D schematic of the memristive devices used to build the 2D axisymmetric model in COMSOL for the SET and RESET processes. Leveraging a 2D geometry in COMSOL facilitates the reduction of volume integrals in equation (1) to area integrals. The area integrals directly corresponding to the SET and RESET processes in COMSOL are expressed as [47-48]:

$$F_{\text{SET}} = \iint \rho C_P \delta T dr dz + \frac{1}{2} \iint \varepsilon |E|^2 dr dz + 2\pi r h \sigma_S + \pi r^2 h \delta \mu_1$$
 (2)

$$F_{\text{RESET}} = \iint \rho C_P \delta T dr dz + \frac{1}{2} \iint \varepsilon |E|^2 dr dz + 2\pi r h \sigma_S + \pi r^2 l \delta \mu_2$$
 (3)

In COMSOL modelling, the values of electric field (E) and temperature (T) can be determined by solving a set of partial differential equations, as presented below:

[i] Electric current module:

$$\nabla . J = 0 \tag{4.1}$$

$$J = \sigma E \tag{4.2}$$

$$E = -\nabla V \tag{4.3}$$

[ii] Heat transfer module (in solid):

$$-\kappa \nabla^2 T = Q_{\rm S} \tag{5}$$

[iii] Multiphysics module:

$$Q_{S} = J.E \tag{6}$$

Herein, J: current density, σ : electric conductivity, κ : thermal conductivity and Q_s : heat source. These equations are typically integrated into their corresponding COMSOL modules and extended for numerical modelling purposes.

Equations (4.1 to 4.3) establish the current conservation law, Ohm's law, and the relationship between electric field and electric potential derived from Maxwell's law, respectively. Equation (5) represents the Fourier heat law, with the Joule heat term from equation (6) providing the heat source. Detailed descriptions of the boundary conditions and electrical configurations employed during modelling are provided elsewhere [50].

In this modelling approach, internal Joule heating and non-uniform electric field distribution within the memristive device are taken into account. Additionally, the study distinctly illustrates how the number of MoS₂ layers impacts the device's switching voltage and synaptic weight, characterized by potentiation (P) and depression (D), which constitutes the core concept of the research. The coefficients of the differential equations, including (4.1) to (4.3), (5), and (6), as well as the free energy equations (2) and (3) employed in this physical electro-thermal modelling, are provided in Table I. Herein, the phonon assisted hopping mechanism is used through the optimum hopping chains. Additionally, the material non-crystallinity is one of the most common features of filamentary RRAM structures. In a noncrystalline material, some atoms or groups of atoms retain a certain mobility being able to move between two equilibrium positions which is described in terms of double well potential (DWP) as discussed in our previous report [50].

Table I: Values of the coefficients of the differential equations and FE used in this physical electro-thermal modelling.

Materials	Electrical Condu	ctivity (σ) [S/m]	Thermal Conductivity (κ) [W/K.m]	Specific Heat Capacity [J/kg. K]	Relative Permittivity (ε_r)	Mass Density (ρ) [kg/m³]
SiO_2	1×10 ³		1.38	703	3.9	2.20×10^{3}
Al	3.8×10 ⁷		235	904	-∞	2.70×10^{3}
MoS_2	1×10 ⁴ [51]		130 [52]	29.2	3.7	5.06×10^{3}
Y_2O_3	10 ⁻¹¹ [53]		0.3 [54]	440	15	5.01×10^{3}
Y_2O_{3-x}	$\sigma_{\rm if} \exp\left(\sigma_{\rm f} \ln\left(\frac{t}{t_{\rm i}}\right)\right) \exp\left(\sqrt{\frac{eV}{kT}}\right)$		$\sigma_{\rm c}(T,V)TL$	528	-∞	6.01×10 ³
Gap	$\sigma_{\rm ig} exp\left(-\sigma_{\rm g} ln\left(\frac{t}{t_{\rm i}}\right)\right) exp\left(\sqrt{\frac{eV}{kT}}\right)$		$k_{ m eff}\sigma_{ m c}(T,V)TL$	440	15	5.01×10 ³
	Parameters	Values		Parameters	Values	
	$\sigma_{ m if}$	5 kS/m	1	$R_{ m L}$	3 kΩ	1
	$\sigma_{ m ig}$	3 kS/m	Circuitry	$V_{(+)}$	1 V	1
	$\alpha_{ m f}$	-0.05		V (-)	-1.5 V	1

Electrical Conductivity	$lpha_{ m g}$	0.05		λ	100 V/s, 10 kV/s, 1 MV/s
	t	V/ λ	Thermal	$k_{ m eff}$	10
			Conductivity (Gap)	0.1	
	t.	0.1ns			

3. RESULTS and DISCUSSION

Figure 2(a) illustrates the simulated RS behaviour of the 2D MoS₂ integrated thin Y₂O₃-based memristive device subjected to a voltage pulsing scheme in the range -1.5 V to +1 V in forward and reverse bias, delivered to the top electrode through a load resistance. The RS response is segmented into four phases: positive forming voltage $(+V_{\rm F})$, positive SET voltage $(V_{\rm SET})$, negative rupture voltage ($-V_R$), and negative RESET voltage (V_{RESET}). During the formation process, all CFs are created within the MoS₂ and Y₂O₃ layer of the RS device, causing it to transition into the SET state when subjected to a voltage amplitude equal to or less than $V_{\rm F}$ (as illustrated in Fig. 2(a)). For the single layer MoS₂ integrated Y₂O₃-based memristive device, V_F and V_{SET} are modelled to be +0.81 V and +0.91 V, respectively. Conversely, during the rupturing process, all previously formed CFs are disrupted, leading the memristive device to switch into the RESET state when exposed to a voltage amplitude equal to or less than $-V_R$. The modelled values for $-V_R$ and V_{RESET} are -1.5 V and -0.89 V, respectively, while in the case of double layer MoS₂ with thin Y₂O₃ (shown in Fig. 2(b)), the V_F is +0.83 V, V_{SET} is 0.62 V, $-V_R$ is -1.18 V and V_{RESET} is -0.61 V. Subsequently, in triple layer MoS₂ (shown in Fig. 2(c)), the V_F is +0.88 V, V_{SET} is 0.15 V, - V_R is -0.81 V and V_{RESET} is -0.34 V. From Fig. 3(a-b) it is concluded that the SET and RESET voltage values decrease as the number of 2D MoS2 monolayers increases. This proves that the presence of 2D MOS₂ plays a pivotal role in the switching process, as compared to primary metal oxide layer. It is well known that the 2D MoS₂ layer requires lower bias potential to switch the device in SET and RESET region, because it shows quantum confinement effect and high carrier mobility [55]. Therefore, charge carriers can move more easily within the material under the influence of a lower electric field, facilitating faster switching at lower bias potentials. This attribute further enhances the low power computation capability of the presented memristive device structure [56].

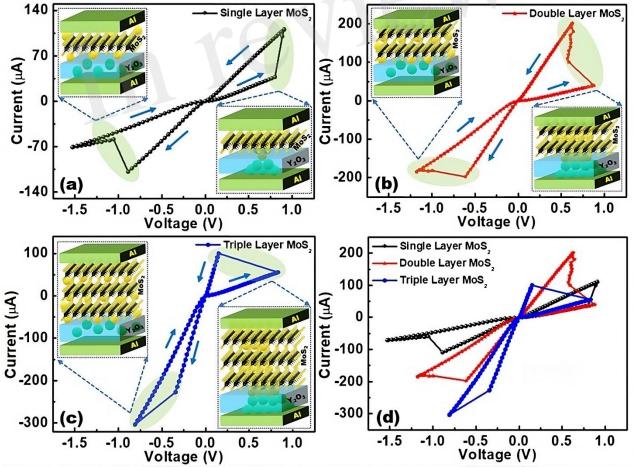


Fig. 2: I-V characteristics depicting resistive switching response of the 2D MoS_2 integrated thin Y_2O_3 -based memristive device for (a) single layer MoS_2 , (b) double layer MoS_2 , (c) triple layer MoS_2 , and (d) comparative analysis of resistive switching response with number of MoS_2 layers.

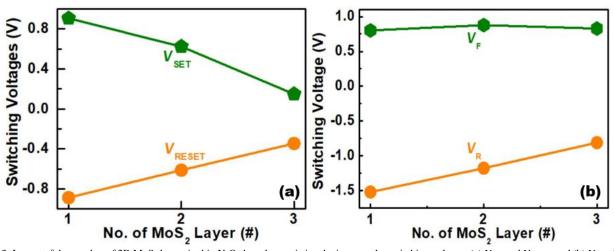


Fig. 3: Impact of the number of 2D MoS₂ layers in thin Y_2O_3 -based memristive device over the switching voltages (a) V_{SET} and V_{RESET} , and (b) V_F and V_R . The lines are guide to the eye.

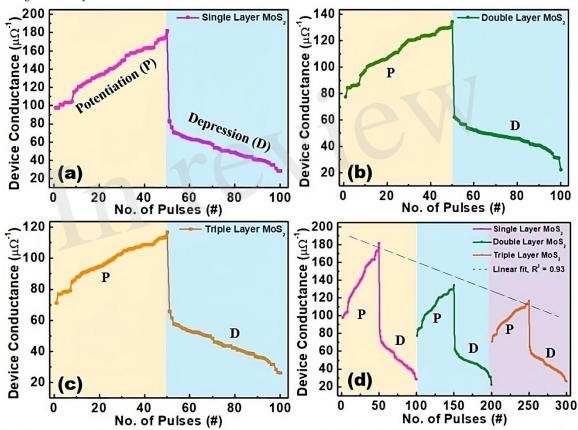


Fig. 4: Impact of the number of 2D MoS_2 layers in thin Y_2O_3 -based memristive device over the device conductance (i.e., synaptic weight) for (a) single layer MoS_2 , (b) double layer MoS_2 , (c) triple layer MoS_2 , and (d) comparative analysis in the change of device conductance with number of MoS_2 layers.

Additionally, to demonstrate the synaptic plasticity characteristics of the memristive device, such as potentiation and depression, a series of positive and negative voltage pulses with an amplitude of ± 1 V and a voltage rise rate (V_{RR}) of 100 V/s are administered to the device. Fig. 4 shows the potentiation and depression functionality of single layer MoS₂ (Fig. 4(a)), double layer MoS₂ (Fig. 4(b)) and triple layer MoS₂ (Fig. 4(c)). As depicted in Fig. 4(a), when subjected to positive voltage pulses, the synaptic weight or normalized conductance of the memristive device undergoes a continuous reinforcement. Conversely, in response to negative electrical stimuli the synaptic weight

diminishes. This ongoing modulation of the device's conductance closely mimics the synaptic plasticity mechanisms observed in the brain [57-58].

As evident from Fig. 4(d), the synaptic weight (or device conductance) of the memristive device is linearly decreased with an increment in the number of MoS₂ layers within the device structure. It should be noted that in monolayer (~ 0.65 nm thickness) MoS₂, quantum confinement effects are more pronounced due to the atomic layer thickness of the material, resulting in enhanced carrier mobility [55]. However, as the number of layers increases, these effects diminish, leading to reduced carrier mobility as well as conductivity.

Additionally, this trend may be attributed to alterations in the lateral dimensions of the device. Moreover, in configurations with a higher number of MoS₂ layers, the role of the thin metal oxide layer in the switching process diminishes, with primary switching being predominantly influenced by the MoS₂ layers rather than the Y₂O₃ layer, which basically constitutes combination of interfacial and filamentary switching mechanism[59-62]. This shift dominance significantly impacts the overall conductance of the device. In this study, a series of 100 identical pulses (comprising 50 positive and 50 negative pulses) with an amplitude of ± 1 V and a pulse width of 10 ms are employed to analyse the synaptic characteristics. Consistent with findings in existing literature [63-66], the metal/metal oxide interfaces demonstrate a gradual potentiation process and an abrupt depression process, as illustrated in Fig. 4(d). The sudden shift observed during the depression process is attributed to variations in the free energy at metal/metal oxide interfaces [67]. Additionally, accounting for the metal/metal oxide free energy and induced oxide layer, non-identical spike pulses featuring varying pulse amplitudes or durations offer a potential solution to

mitigate the high asymmetry ratio observed in potentiation and depression processes [67]. However, employing non-identical spikes may introduce complexity to peripheral circuits and neuro-inspired computing systems [67].

Fig. 5 depicts the impact of pulse amplitude on synaptic plasticity, delineating both potentiation and depression processes. The graphs illustrate how altering the pulse amplitude effectively modulates the device conductance for single (Fig. 5(a)), double (Fig. 5(b)) and triple layer MoS₂ (Fig. 5(a)), akin to the way spikes in neural communication influence synaptic strength. Notably, successive potentiation spikes elevate memristive conductance, while subsequent depression spikes induce a cycle of conductance reduction. The different pulsing scheme (non-identical pulses) is utilized during the computation of Fig. 5. Maldonado et al [8] have observed similar behaviour experimentally in HfO2-based devices. Therefore, considering memristive aforementioned analysis, it can be concluded that control over the MoS₂ number of layers affects the conductance but not the synaptic response of the Yttria-based memristive device.

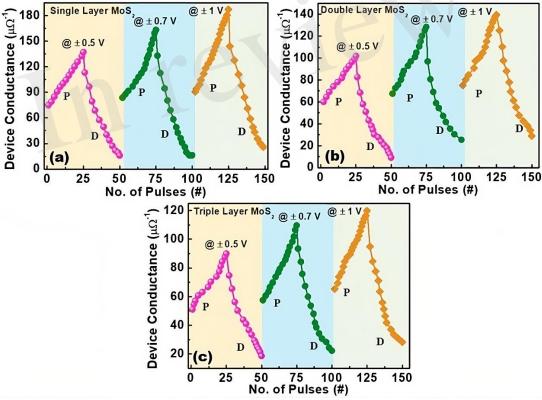


Fig. 5: Synaptic plasticity, potentiation, and depression events in the memristive device under the application of varying pulsing scheme (non-volatile states) for (a) single layer MoS_2 , (b) double layer MoS_2 , and (c) triple layer MoS_2 .

4. CONCLUSION

We presented a physical model detailing the integration of a number of nanoscales 2D MoS_2 layers with a thin metal oxide-based memristive device, aimed at emulating artificial synapse functionalities. The simulations demonstrate a bipolar RS response across all different number of MoS_2 layers, with device switching

voltages decreasing as the number of MoS₂ layers increases. This phenomenon enhances the feasibility of 2D materials combined with transition metal oxide-based memristive devices in low-power computing applications. Moreover, the MoS₂-modified memristive device structure successfully demonstrated synaptic plasticity in terms of potentiation and depression, while the device conductance can also be varied by tuning the amplitude of the input pulsing scheme. Notably, in

monolayer MoS₂, quantum confinement effects are more dominant due to the atomic layer thickness of the material, resulting in enhanced carrier mobility and corresponding increment in the device conductance. Conversely, in double and triple-layer MoS₂, these effects are minimized, which further affects the synaptic response of the device. However, in double and tri-layer device structures, the bulk materials conductivity may play a more important role, affecting thus the device switching voltages more significantly, as compared to the overall effect on device conductance. Moreover, the modified memristive device structure with the integration of 2D MoS₂ can pave the way for optoelectronic memories development that may also act as optoelectronic synapses in bionic visual applications. Hence, the profound advantage of the herein presented physical model lies in its ability to inform researchers about the potential functionalities of MoS2 integrated Y₂O₃-based memristive devices and guide them in developing novel low computing power 2D-TMD/TMObased artificial synapses for tuneable neuromorphic computing applications.

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REFERENCES

- [1] Q. Xia, J. J. Yang, "Memristive crossbar arrays for brain-inspired computing", Nature Mater, vol. 18, pp. 309-323, 2019, DOI: 10.1038/s41563-019-0291-x.
- [2] J. J. Yang, D. B. Strukov, D. R. Stewart, "Memristive devices for computing", Nature Nanotech, vol. 8, pp. 13-24, 2013, DOI: 10.1038/nnano.2012.240.
- [3] M. A. Zidan, J. P. Strachan, W. D. Lu, "The future of electronics based on memristive systems", Nature Electronics, vol. 1, pp. 22-29, 2018, DOI: 10.1038/s41928-017-0006-8.
- [4] Z. Wang, S. Joshi, S. E. Savel'ev, H. Jiang, R. Midya, P. Lin, M. Hu, N. Ge, J. P. Strachan, Z. Li, Q. Wu, M. Barnell, G. L. Li, H. L. Xin, R. S. Williams, Q. Xia and J. J. Yang, "Memristors with diffusive dynamics as synaptic emulators for neuromorphic computing", Nature Mater, vol. 16, pp. 101-108, 2017, DOI: 10.1038/nmat4756.
- [5] H. Sato, H. Shima, T. Nokami, T. Itoh, Y. Honma, Y. Naitoh, H. Akinaga, K. Kinoshita, "Memristors with Controllable Data Volatility by Loading Metal Ion-Added Ionic Liquids", Frontiers in Nanotechnology, vol. 3, 2021, DOI: 10.3389/fnano.2021.660563.
- [6] A. Alshaya, Q. Han, C. Papavassiliou, "RRAM, Device, Model and Memory", International Conference on Microelectronics (ICM), 2022. DOI: 10.1109/ICM56065.2022.10005367.
- [7] F. Raffone, "Physical processes and materials in memristive devices: a theoretical study", 2017.

- [8] D. Maldonado, A. Cantudo, E. Perez, R. R. Zaliz, E. P. B. Quesada, M. K. Mahadevaiah, F. J. Molinos, C. Wenger, and J. B. Roldan, "TiN/Ti/HfO₂/TiN memristive devices for neuromorphic computing: from synaptic plasticity to stochastic resonance", Front. Neurosci. vol. 17, no. 1271956, 2023, DOI: 10.3389/fnins.2023.1271956.
- [9] S. Kumar, D. D. Kumbhar, J. H. Park, R. K. Kamat, T. D. Dongale and S. Mukherjee, "Y₂O₃-Based Crossbar Array for Analog and Neuromorphic Computation", IEEE Transactions on Electron Devices, vol. 70, no. 2, 2022, pp. 473-477, DOI: 10.1109/TED.2022.3227890.
- [10] S. Kumar, A. Agarwal, and S. Mukherjee, "Electrical Performance of Large-area Y₂O₃ Memristive Crossbar Array with Ultralow C2C Variability", IEEE Transactions on Electron Devices, vol. 69, no. 7, 2022, pp. 3660-3666, DOI: 10.1109/TED.2022.3172400.
- [11] S. Kumar, M. Das, M. T. Htay, S. Sriram, and S. Mukherjee, "Electroforming-free Y_2O_3 Memristive Crossbar Array with Low Variability", ACS Applied Electronic Materials, vol. 4, no. 6, 2022, pp. 3080-3087, DOI: 10.1021/acsaelm.2c00472.
- [12] S. Kumar, M. K. Gautam, S. Yadav, and S. Mukherjee, "Memcapacitive to Memristive Transition in Al/Y₂O₃/GZO Crossbar Array", IEEE Transactions on Electron Devices, vol. 70, no. 6, 2023, pp. 3341-3346, DOI: 10.1109/TED.2023.3265622.
- [13] M. K. Gautam, S. Kumar, S. Chaudhary, L. K. Hindoliya, D. D. Kumbhar, J. H. Park, M. T. Htay, and S. Mukherjee, "Experimental Validation of Switching Dependence of Nanoscale-Y₂O₃ Memristor on Electrode Symmetry via Physical Electro-Thermal Modeling", ACS Applied Electronic Materials, vol. 5, no. 7, 2023, pp. 3885-3893, DOI: 10.1021/acsaelm.3c00598.
- [14] D. D. Kumbhar, S. Kumar, M. Dubey, A. Kumar, T. D. Dongale, S. D. Pawar, and S. Mukherjee, "Exploring Statistical Approaches for Accessing the Reliability of Y₂O₃-based Memristive Devices", Microelectronic Engineering, vol. 288, no. 112166, 2024, DOI: 10.1016/j.mee.2024.112166.
- [15] M. Kim, M. A. Rehman, D. Lee, Y. Wang, D. H. Lim, M. F. Khan, H. Choi, Q. Y. Shao, J. Suh, H. S. Lee, and H. H. Park, "Filamentary and Interface-Type Memristors Based on Tantalum Oxide for Energy-Efficient Neuromorphic Hardware", ACS Appl. Mater. Interfaces, vol. 14, no. 39, pp. 44561-44571, 2022, DOI: 10.1021/acsami.2c12296.
- [16] D. B. Strukov, G. S. Snider, D. R. Stewart, and R. S. Williams, "The missing memristor found" Nature, vol. 453, pp. 80-83, 2008, DOI: 10.1038/nature06932.[17] M. Naqi, M. S. Kang, N. liu, T. Kim, S. Baek, A. Bala, C. Moon, J. Park, and S. Kim, "Multilevel artificial electronic synaptic device of direct grown robust MoS₂ based memristor array for in-memory deep neural network", npj 2D Mater Appl, vol. 6, no. 53, 2022, DOI: 10.1038/s41699-022-00325-5.
- [18] Q. Cao, L. Xiong, X. Yuan, P. Li, J. Wu, H. Bi, and J. Zhang, "Resistive switching behavior of the memristor based on WS_2 nanosheets and polyvinylpyrrolidone nanocomposites", Appl. Phys. Lett., vol. 120, no. 232105, 2022, DOI: 10.1063/5.0087862
- [19] H. Duan, D. Wang, J. Gou, F. Guo, W. Jie, and J. Hao, "Memristors based on 2D MoSe₂ nanosheets as artificial synapses and nociceptors for neuromorphic computing", Nanoscale, vol. 15, pp. 10089-10096, 2023, DOI: 10.1039/D3NR01301D.
- [20] R. Ge, X. Wu, M. Kim, J. Shi, S. Sonde, L. Tao, Y. Zhang, J. C. Lee, and D. Akinwande, "Atomristor: nonvolatile resistance switching in atomic sheets of transition metal dichalcogenides", Nano Lett, vol. 18, pp. 434-441, 2018, DOI: 10.1021/acs.nanolett.7b04342.
- [21] M. Kim, R. Ge, X. Wu, X. Lan, J. Tice, J. C. Lee, and D. Akinwande, "Zero-static power radio-frequency switches based on MoS_2 atomristors. Nat Commun, vol. 9, no. 2524, 2018, DOI: 10.1038/s41467-018-04934-x.
- [22] R. Luo, W. W. Xu, Y. Zhang, Z. Wang, X. Wang, Y. Gao, P. Liu, and M. Chen, "Van der Waals interfacial reconstruction in monolayer transition-metal dichalcogenides and gold heterojunctions. Nature Communications, vol. 11, no. 1, 2020, DOI: 10.1038/s41467-020-14753.8
- [23] E. A. Marseglia, "Transition Metal Dichalcogenides and Their Intercalates", International Reviews in Physical Chemistry, vol. 3, no. 2, 1983, DOI: 10.1080/01442358309353343.
- [24] X. Yin, C. S. Tang, D. Wu, W. Kong, C. Li, Q. Wang, L. Cao, M. Yang, Y. H. Chang, D. Qi, F. Ouyang, S. J. Pennycook, Y. P. Feng, M.

- B. H. Breese, S. J. Wang, W. Zhang, A. Rusydi, A. T. S. Wee "2D Transition Metal Dichalcogenide: Unraveling High-Yield Phase-Transition Dynamics in Transition Metal Dichalcogenides on Metallic Substrates, vol. 6, no. 7, 2019, DOI: 10.1002/advs.201970042.
- [25] X. F. Lu, Y. Zhang, N. Wang, S. Luo, K. Peng, L. Wang, H. Chen, W. Gao, X. H. Chen, Y. Bao, G. Liang, and K. P. Loh, "Exploring low power and ultrafast memristor on p-type van der Waals SnS", Nano Lett., vol. 21, no. 20, pp. 8800-8807, 2021, DOI: 10.1021/acs.nanolett.1c03169.
- [26] S. Li, M. E. Pam, Y. Li, L. Chen, Y. C. Chien, X. Fong, D. Chi, K. W. Ang, "Wafer-scale 2D hafnium diselenide based memristor crossbar array for energy-efficient neural network hardware", Adv Mater, vol. 34, no. 2103376, 2022, DOI: 10.1002/adma.202103376.
- [27] K. Ranganathan, M. F. Raz, A. Ismach, "Large-scale and robust multifunctional vertically aligned MoS₂ photomemristors", Adv Funct Mater, vol. 30, no. 2005718, 2020, DOI: 10.1002/adfm.202005718.
- [28] X. Feng, Y. Li, L. Wang, S. Chen, Z. G. Yu, W. C. Tan, N. Macadam, G. Hu, L. Huang, L. Chen, X. Gong, D. Chi, T. Hasan, A. V. Y. Thean, Y. W. Zhang, K. W. Ang, "A fully printed flexible MoS_2 memristive artificial synapse with femtojoule switching energy", Adv Electron Mater, vol. 5, no. 1900740, 2019, DOI: 10.1002/aelm.201900740.
- [29] X. Yan, Q. Zhao, A. P. Chen, J. Zhao, Z. Zhou, J. Wang, H. Wang, L. Zhang, X. Li, Z. Xiao, K. Wang, C. Qin, G. Wang, Y. Pei, H. Li, D. Ren, J. Chen, and Q. Liu, "Vacancy-induced synaptic behavior in 2D WS $_2$ nanosheet-based memristor for low-power neuromorphic computing", Small, vol. 15, no. 1901423, 2019, DOI: 10.1002/smll.201901423.
- [30] W. Tong, and Y. Liu, "Recent progress of layered memristors based on two-dimensional MoS₂", Science China, vol. 66, pp. 160402:1-160402:10, 2023, DOI: 10.1007/s11432-023-3751-y.
- [31] V. K. Sangwan, D. Jariwala, I. S. Kim, K. S. Chen, T. J. Marks, L. J. Lauhon, and M. C. Hersam, "Gate-tunable memristive phenomena mediated by grain boundaries in single-layer MoS₂. Nat Nanotech, vol. 10, pp. 403-406, 2015, DOI: 10.1038/nnano.2015.56.
- [32] M. D. Pickett, D. B. Strukov, J. L. Borghetti, J. J. Yang, G. S. Snider, D. R. Stewart, R. S. Williams, "Switching dynamics in titanium dioxide memristive devices", Journal of Applied Physics, vol. 106, no. 7, 2009, DOI: 10.1063/1.3236506.
- [33] Y. Wang, Y. Gong, S. Huang, X. Xing, Z. Lv, J. Wang, J. Q. Yang, G. Zhang, Y. Zhou, and S. T. Han, "Memristor-based biomimetic compound eye for real-time collision detection", Nature Communications, vol. 12, no. 5979, 2021, DOI: 10.1038/s41467-021-26314-8.
- [34] Y. Lin, W. Wang, R. Li, J. Kim, C. Zhang, H. Kan, and Y. Li, "Multifunctional optoelectronic memristor based on CeO₂/MoS₂ heterojunction for advanced artificial synapses and bionic visual system with nociceptive sensing", Nano Energy, vol. 121, no. 109267, 2024, DOI: 10.1016/j.nanoen.2024.109267.
- [35] R. Li, W. Wang, Y. Li, S. Gao, W. Yue, and G. Shen, "Multimodulated optoelectronic memristor based on Ga_2O_3/MoS_2 heterojunction for bionic synapses and artificial visual system", Nano Energy, vol. 111, no. 108398, 2023, DOI: 10.1016/j,nanoen.2023.108398.
- [36] S. Kim, S. J. Kim, K. M. Kim, S. R. Lee, M. Chang, E. Cho, Y. B. Kim, C. J. Kim, U. I. Chung, and I. K. Yoo, "Physical electro-thermal model of resistive switching in bi-layered resistance-change memory", Sci. Rep., vol. 3, no. 1680, 2013, DOI: 10.1038/srep01680.
- [37] M. Bocquet, D. Deleruyelle, H. Aziza, C. Muller, J. M. Portal, T. Cabout, and E. Jalaguier, "Robust compact model for bipolar oxide-based resistive switching memories," IEEE Trans. Electron Devices, vol. 61, no. 3, 2014, DOI: 10.1109/TED.2013.2296793.
- [38] B. Traore, P. Blaise, E. Vianello, L. Perniola, B. D. Salvo, and Y. Nishi, "HfO₂-based RRAM: Electrode effects, Ti/HfO₂ interface, charge injection, and oxygen (O) defects diffusion through experiment and ab initio calculations," IEEE Trans. Electron Devices, vol. 63, no. 1, 2016, DOI: 10.1109/TED.2015.2503145.
- [39] S. Kumar, M. K. Gautam, G. S. Gill, and S. Mukherjee, "3-D Physical Electro-Thermal Modelling of Nanoscale Y₂O₃ Memristors for Synaptic Application", IEEE Transactions on Electron Devices, vol. 69, no. 6, 2022, pp. 3124-3129, DOI: 10.1109/TED.2022.3166858.
- [40] G. Korotcenkov, and B.K. Cho, "The role of grain size on the thermal instability of nanostructured metal oxides used in gas sensor

- applications and approaches for grain-size stabilization", vol. 58, no. 4, 2012, pp. 167-208, DOI: 10.1016/j.pcrysgrow.2012.07.001.
- [41] D. Ielmini, "Resistive switching memories based on metal oxides: mechanisms, reliability and scaling", Semicond. Sci. Technol., vol. 31, no. 6, 2016, DOI: 10.1088/0268-1242/31/6/063002.
- [42] S. Kim, S. J. Kim, K. M. Kim, S. R. Lee, M. Chang, E. Cho, Y. B. Kim, C. J. Kim, U. I. Chung, and I. K. Yoo, "Physical electro-thermal model of resistive switching in bi-layered resistance-change memory," Sci. Rep., vol. 3, no. 1680, 2013, DOI: 10.1038/srep01680.
- [43] B. Peng, H. Zhang, H. Shao, Y. Xu, X. Zhang and H. Zhu, "Thermal conductivity of monolayer MoS₂, MoSe₂, and WS₂: interplay of mass effect, interatomic bonding and anharmonicity", RSC Adv., vol. 6, pp. 5767-5773, 2016, DOI: 10.1039/C5RA19747C.
- [44] X. F. Wang, H. Tian, H. M. Zhao, T. Y. Zhang, W. Q. Mao, Y. C. Qiao, Y. Pang, Y. X. Li, Y. Yang, T. L. Ren, "Interface Engineering with MoS₂-Pd Nanoparticles Hybrid Structure for a Low Voltage Resistive Switching Memory", Small, vol. 14, no. 2, 2018, DOI: 10.1002/smll.201702525.
- [45] F. Wu, S. Si, P. Cao, W. Wei, X. Zhao, T. Shi, X. Zhang, J. Ma, R. Cao, L. Liao, T. Y. Tseng, and Q. Liu, "Interface Engineering via MoS₂ Insertion Layer for Improving Resistive Switching of Conductive-Bridging Random Access Memory", Advanced Electronic Materials, vol. 5, no. 4, 2019, DOI: 10.1002/aelm.201800747.
- [46] S. Jin, J. D. Kwon, Y. Kim, S. Jin, J. D. Kwon, Y. Kim, "Statistical Analysis of Uniform Switching Characteristics of Ta_2O_5 -Based Memristors by Embedding In-Situ Grown 2D-MoS $_2$ Buffer Layers", Materials, vol. 14, no. 21, 2021, DOI: 10.3390/ma14216275.
- [47] D. Niraula and V. Karpov, "Numerical modeling of resistive switching in RRAM device," in Proceedings of the 2017 COMSOL Conference in Boston (COMSOL Inc., 2017), available at https://www.comsol.com/paper/download/437382.
- [48] D. Niraula, and V. Karpov, Comprehensive numerical modeling of filamentary RRAM devices including voltage ramp-rate and cycle-to-cycle variations, J. Appl. Phys., vol. 124, no. 17, 2018, DOI: 10.1063/1.5042789.
- [49] S. Gaba, P. Sheridan, J. Zhou, S. Choi and W. Lu, "Stochastic memristive devices for computing and neuromorphic applications", Nanoscale, 2013, vol. 5, DOI: 10.1039/C3NR01176C.
- [50] S. Kumar, M. Dubey, M. Nawaria, M. K. Gautam, M. Das, R. Bhardwaj, S. Rani, and S. Mukherjee, "Investigation of Filament Formation and Surface Perturbation in Nanoscale-Y₂O₃ Memristor: A Physical Modelling Approach," Journal of Electronic Materials, Springer Nature, 2024, DOI: 10.1007/s11664-024-10967-4.
- [51] O. El Beqqali, I. Zorkani, F. Rogemond, H. Chermette, R. Ben Chaabane, M. Gamoudi, G. Guillaud, Electrical properties of molybdenum disulfide MoS₂: Experimental study and density functional calculation results, Synthetic Metals, vol. 90, no. 3, 1997, pp. 165-172, DOI: 10.1016/S0379-6779(98)80002-7.
- [52] Y. Yu, T. Minhaj, L. Huang, Y. Yu, and L. Cao, "In-Plane and Interfacial Thermal Conduction of Two-Dimensional Transition-Metal Dichalcogenides", Phys. Rev. Applied 13, 034059, 2020, DOI: 10.1103/PhysRevApplied.13.034059.
- [53] https://inis.iaea.org/search/search.aspx?orig_q=RN:33056693.
- [54] https://www.makeitfrom.com/material-properties/Yttria-Yttrium-Oxide- Y_2O_3 .
- [55] Y. Ling, J. Li, T. Luo, Y. Lin, G. Zhang, M. Shou and Q. Liao, "MoS₂-Based Memristor: Robust Resistive Switching Behavior and Reliable Biological Synapse Emulation", Nanomaterials, vol. 13, no. 3117, 2023, DOI: 10.3390/nano13243117
- [56] R. Xu, H. Jang, M. H. Lee, D. Amanov, Y. Cho, H. Kim, S. Park, H. J. Shin, and D. Ham, "Vertical MoS₂ Double-Layer Memristor with Electrochemical Metallization as an Atomic-Scale Synapse with Switching Thresholds Approaching 100 mV", Nano Lett. 2019, 19, 4, 2411-2417 2019, DOI: 10.1021/acs.nanolett.8b05140
- [57] S. H. Jo, T. Chang, I. Ebong, B. Bhadviya, P. Mazumder and W. Lu, "Nanoscale memristor device as synapse in neuromorphic systems", Nano Lett., vol. 10, no. 4, 2010, DOI: 10.1021/nl904092h.
- [58] M. Das, A. Kumar, R. Singh, M. T. Htay and S. Mukherjee, "Realization of synaptic learning and memory functions in Y_2O_3 based memristive device fabricated by dual ion beam sputtering", Nanotechnology, vol. 29, no. 055203, pp. 1-9, 2018, DOI: 10.1088/1361-6528/aaa0eb.
- [59] M. E. Pam, S. Li, T. Su, Y. C. Chien, Y. Li, Y. S. Ang, K. W. Ang, "Interface-Modulated Resistive Switching in Mo-Irradiated ReS₂ for

Neuromorphic Computing", Advanced Materials, vol. 34, no. 30, 2022, DOI: 10.1002/adma.202202722.

[60] S. Ginnaram, and S. Maikap, "Memristive and artificial synapse performance by using TiO_x/Al_2O_3 interface engineering in MoS_2 -based metallic filament memory", Journal of Physics and Chemistry of Solids, vol.151, 2021, DOI: 10.1016/j.jpcs.2020.109901.

[61] W. Wang, G. N. Panin, X. Fu, L. Zhang, P. Ilanchezhiyan, V. O. Pelenovich, D. Fu, and T. W. Kang, "MoS₂ memristor with photoresistive switching", Scientific Reports, vol. 6, no. 31224, 2016, DOI: 10.1038/srep31224.

[62] D. Li, B. Wu, X. Zhu, J. Wang, B. Ryu, W. D. Lu, W. Lu, X. Liang, "MoS₂ Memristors Exhibiting Variable Switching Characteristics toward Biorealistic Synaptic Emulation", ACS Nano, vol. 12, no. 9, 2018, DOI: 10.1021/acsnano.8b03977.

[63] J. W. Jang, S. Park, Y. H. Jeong and H. Hwang, "ReRAM-based Synaptic Device for Neuromorphic Computing", 2014 IEEE International Symposium on Circuits and Systems (ISCAS),

Melbourne, VIC, Australia, no. 14483574, 2014, DOI: 10.1109/ISCAS.2014.6865320.

[64] H. Liu, M. Wei and Y. Chen, "Optimization of non-linear conductance modulation based on metal oxide memristors", Nanotechnol Rev., vol. 5, no. 7, 2018, DOI: 10.1515/ntrev-2018-0045. [65] K. Kim, S. Park, S. M. Hu, J. Song, W. Lim, Y. Jeong, J. Kim, S. Lee, J. Y. Kwak, J. Park, J. K. Park, B. K. Ju, D. S. Jeong, and I. Kim, "Enhanced analog synaptic behavior of SiN_x/a-Si bilayer memristors through Ge implantation", NPG Asia Materials, vol. 12, no. 77, 2020, DOI: 10.1038/s41427-020-00261-0.

[66] J. J. Zhang, H. J. Sun, Y. Li, Q. Wang, X. H. Xu, and X. S. Miao, "AgInSbTe memristor with gradual resistance tuning", Appl. Phys. Lett., vol. 102, no. 18,2013, DOI: 10.1063/1.4804983

[67] S. Yu, "Neuro-inspired computing using resistive synaptic devices", Springer: Berlin, 2017, ISBN 978-3-319-54313-0, DOI: 10.1007/978-3-319-54313-0.



