High-uniformity Memristor Arrays Based on Two-dimensional MoTe₂ for Neuromorphic Computing

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Abstract: Two-dimensional transition metal dichalcogenides are appealing materials for the preparation of nanoelectronic devices, and the development of memristors for information storage and neuromorphic computing using such materials is of particular interest. However, memristor arrays based on two-dimensional transition metal dichalcogenides are rarely reported due to low yield and high device-to-device variability. Herein, the memristive devices based on centimeter-scale two-dimensional MoTe₂ film are firstly reported. The 2D MoTe₂ film was prepared by the chemical vapor deposition method. Then the memristive devices based on 2D MoTe₂ film were fabricated through the polymethyl methacrylate transfer method and the lift-off process. The prepared MoTe₂ devices perform stable bipolar resistive switching, including superior retention characteristics (>1000 s), fast switching (~60 ns for SET and ~280 ns for RESET), and excellent endurance (>2000 cycles). More importantly, the MoTe₂ devices exhibit high yield (96%), low cycle-to-cycle variability (6.6% for SET and 5.2% for RESET), and low device-to-device variability (19.9% for SET and 15.6% for RESET). In addition, a 3×3 memristor array with 1R scheme is first and successfully demonstrated based on 2D MoTe₂ film. And, high recognition accuracy (91.3%) has been realized by simulation in the artificial neural network with the MoTe₂ devices working as synapses. It is found that the formation/rupture of metallic filaments is the dominating switching mechanism based on the investigations of the electron transport characteristics of high and low resistance states in the present MoTe₂ devices. This work demonstrates that large-scale two-dimensional transition metal dichalcogenides film is of great potential for future applications in neuromorphic computing.

Key words: two-dimensional materials; MoTe₂; memristor array; neuromorphic computing

The last few years have witnessed rapid progress towards the realization of artificial intelligence (AI), especially software AI thanks to the advances in the development of the algorithm. Note that software AI is typically demonstrated based on the digital computer with conventional Von Neumann architectures at the cost of huge power consumption and massive data throughput[11]. In contrast, hardware AI systems based on in-memory computing architecture can handle probabilistic and unstructured problems with low power dissipation resembling biological neural networks. Recently, memristor has attracted increasing attention as a promising candidate for the construction of hardware neuromorphic computing systems due to its prominent advantages, including simple structure and rich switching dynamics resembling biological synapses and neurons[2-8].

Emerging two-dimensional (2D) materials, especially 2D layered transition metal dichalcogenides (TMDs), are actively studied for fabricating high-performance nano electronic devices[9-11]. The 2D TMDs have shown prospective potential for memristor
applications, and these devices could be of use in both information storage and neuromorphic computing\cite{12}. Such devices exhibit properties that traditional thin film-based memristors do not have, including high thermal stability\cite{13}, high controllability of potentiation, depression and relaxation\cite{14}, excellent flexibility and transparency\cite{15}. However, memristors based on 2D TMDs are typically fabricated by mechanical exfoliation, which is not feasible for large-scale array preparation; thus the yield and device-to-device variability of 2D TMD-based devices are rarely reported.

Herein, the memristive devices were prepared based on 2D MoTe$_2$ film fabricated by chemical vapor deposition (CVD). This MoTe$_2$ device shows stable bipolar resistive switching with superior retention characteristics, good endurance, high yield, and excellent uniformity. Furthermore, without selector devices, a 3×3 memristor array was first demonstrated based on the MoTe$_2$ film. And, a handwritten digits recognition neural network simulation was implemented using the prepared MoTe$_2$ devices as synapses. This work indicates that large-scale 2D TMDs films are promising materials for future neuromorphic computing.

1 Experimental

1.1 Device fabrication

2D MoTe$_2$ film was prepared through the CVD method. Firstly, a 1 nm Mo film was deposited onto a heavily doped Si substrate with 300 nm SiO$_2$ by electron beam evaporation. Then the Mo film was fully oxidized to MoO$_3$ in air. The resulting MoO$_3$ film was placed in a ceramic crucible containing Te powder. A mixture of argon and hydrogen was used as the carrier gas and formed a reducing atmosphere during the CVD growth. The MoO$_3$ film was tellurized into a MoTe$_2$ film after annealing in Te vapor at 700 °C. The bottom conductive layer (30 nm Au/10 nm Ti) and the top conductive layer (100 nm Au/10 nm Ti) were deposited through DC sputtering and lift-off process. And the MoTe$_2$ film was transferred onto the bottom conductive layer through the polymethyl methacrylate (PMMA) transfer method. This transfer process is shown schematically in Fig. S1).

1.2 Characterization and electrical measurement

The component of the MoTe$_2$ film was measured by Raman spectra with an inVia Reflex spectrometer operated under a 532 nm laser and the XRD measurement (Bruker D8 Advance). An optical microscope (DSX 510) was employed to check the structure of the Au/Ti/MoTe$_2$/Au/Ti device. The thickness and the structure of the memristor array were measured through an atomic force microscope (AFM, SPM9700). All electrical measurements were conducted in air at room temperature with a Keithley 4200 semiconductor characterization system connected with a tabletop cryogenic probe station (PS-100, Lakeshore).

2 Results and discussion

2.1 Composition and structure characterization of the MoTe$_2$ device

Fig. 1(a) shows a typical photo of the MoTe$_2$ film after being transferred on the silicon substrate with bottom electrodes. The film is uniform and continuous across the whole area (∼1 cm), as can be seen from the homogeneous color contrast in the image. And, this thin film is pure MoTe$_2$ with no other phases, which is verified by the XRD measurement (Fig. S2). Raman spectroscopy was further employed to investigate the structure of the prepared MoTe$_2$ film, as shown in Fig. 1(b). The MoTe$_2$ film shows several Raman peaks between 100 and 300 cm$^{-1}$: the out-of-plane A$_{1g}$ mode at ∼171 cm$^{-1}$ and the prominent peak of the in-plane E$_{2g}$ mode at ∼233 cm$^{-1}$. These Raman features coincide with those observed in few-layer MoTe$_2$ with the hexagonal (2H) phase, thus unequivocally identifying the as-grown film as 2H MoTe$_2$\cite{16-17}.

After characterization of the MoTe$_2$ film, the Au/Ti/MoTe$_2$/Au/Ti device was fabricated through the PMMA transfer method and lift-off process, and the detailed fabrication process is shown in Fig. S3. The optical image of 50 Au/Ti/MoTe$_2$/Au/Ti devices is presented in Fig. 1(c).

In addition to the independent MoTe$_2$ device, a 3×3 memristor array was prepared based on the continuous MoTe$_2$ film. The optical image of the 3×3 memristor array is shown in Fig. 1(d). And the atomic force microscope (AFM) image and height profiles of the prepared array (Fig. S4) demonstrate that the MoTe$_2$ film can adapt to the contour morphology of the bottom electrodes, resulting in a conformal coating.

2.2 Memristive behavior of the
**Au/It/MoTe$_2$/Au/it device**

After the electroforming process (Fig. S5), stable bipolar resistive switching was obtained when the voltage was swept between -0.8 V and 1.0 V, as shown in Fig. 2(a). During a continuous bias sweeping from 0 V→1 V→0.8 V→0 V, a pinched hysteresis loop was obtained, in which SET (switching from high resistance state (HRS) to low resistance state (LRS)) occurred at about 0.7 V and RESET (switching from LRS to HRS) happened at about -0.5 V. A compliance current of 3 mA was applied in the SET process to prevent hard breakdown. This hysteresis behavior is reproducible in the successive 20 voltage sweeps, indicating the stability of the switching behavior in the present device.

**Fig. 1 Characterization of MoTe$_2$ film and electrical measurement of Au/Ti/MoTe$_2$/Au/Ti device**

(a) The photo of the centimeter-scale MoTe$_2$ film; (b) Raman spectrum of the MoTe$_2$ film; (c) The optical image of the prepared memristive devices with the structure of Au/Ti/MoTe$_2$/Au/Ti; (d) The optical image of a 3×3 memristor array

**Fig. 2 Stable bipolar resistive switching behavior and retention characteristics of the MoTe$_2$ device**

(a) 20 cycles of I-V curves with a compliance current of 3 mA; (b) Retention characteristics of the HRS and the LRS read at 0.1 V

In addition to stable bipolar switching behavior and good retention characteristic, the MoTe$_2$ device exhibits fast switching and good endurance. Current responses of the MoTe$_2$ device under the SET and RESET pulses are presented in Fig. 3(a-b). It was found that the device could be switched to LRS in about 60 ns and switched back to HRS in about 280 ns. Furthermore, over 2000 switching cycles can be obtained under SET pulse of 1.7 V/700 ns and RESET pulse of -1.2 V/7 μs (see Fig. 3(c)), indicating good endurance of the present MoTe$_2$ device.

**2.3 High yield and excellent uniformity of the MoTe$_2$ device**

The yield and uniformity of the prepared device are systematically studied, since they are crucial for the construction of large-scale memristor arrays. Among 25 prepared devices, 24 devices show stable bipolar resistive switching behavior similar to that shown in Fig. 2(a), indicating a yield of 96%. We statistically analyzed 480 I-V curves collected in 24 devices (detailed results are shown in Fig. S6), and quantified the cycle-to-cycle and the device-to-device variability of the SET voltage ($V_{\text{SET}}$), RESET voltage ($V_{\text{RESET}}$), HRS and LRS by calculating the coefficient of variation ($C_V$) as the standard deviation ($\sigma$) divided by the mean value ($\mu$), in absolute value.$^{[18]}$ The cycle-to-cycle variability in a single device results from the stochastic nature of the switching process, and the device-to-device variability is attributed to inhomogeneity in the samples derived from the fabrication process$^{[19]}$, such as device area and thickness fluctuations, and so on.

The minimum cycle-to-cycle variabilities of $V_{\text{SET}}$ and $V_{\text{RESET}}$ are 6.6% and 5.2% for a given device, respectively, and the device-to-device variabilities of $V_{\text{SET}}$ and $V_{\text{RESET}}$ rise to 19.9% and 15.6%, respectively, when considering all 24 devices (Fig. 4). In addition, the device-to-device variabilities of HRS and LRS are...
16.8% and 12.7%, respectively (Fig. S7). Note that the device-to-device variabilities of $V_{\text{SET}}$ and $V_{\text{RESET}}$ are 6.06% and 29.07%, respectively, for the CVD h-BN device reported recently[20], indicating the uniformity of the present device is comparable to that of the h-BN device. Such excellent uniformity makes the MoTe$_2$ film promising for the construction of large-scale memristor arrays.

**Fig. 3** Fast switching and good endurance of the MoTe$_2$ device
(a) The SET speed under the pulse with the amplitude 1.3 V; (b) The RESET speed of the MoTe$_2$ device under the pulse with the amplitude of -1.0 V; (c) Over 2000 switching cycles obtained by applying SET pulse of 1.7 V/700 ns and RESET pulse of -1.2 V/7 μs

**2.4 The realization of a 3×3 memristor array**

According to the estimation using the one-bit pull-up scheme (detailed results are shown in Fig. S8 and Fig. S9), the maximum array size with the 1R scheme and with the 1S1R scheme is 4×4 and 870×870 (740 kb), respectively. Therefore, a 3×3 array was fabricated to verify the feasibility of the memristor array based on the MoTe$_2$ film. After an electroforming process, stable resistive switching was achieved in the MoTe$_2$ array device, as shown in Fig. 5(a-b). Notably, due to the inevitable issue of leakage current, the resistance values of the MoTe$_2$ array devices are lower than those of the independent MoTe$_2$ devices, especially for the HRS, leading to a smaller switching ratio (detailed results are presented in Fig. S10). These results demonstrate that the MoTe$_2$ film is promising for the construction of memristor array, and the array size can be further enlarged through combing with proper selectors.

**Fig. 4** Cumulative distribution of $V_{\text{SET}}$ and $V_{\text{RESET}}$ of 24 devices

**Fig. 5** Stable resistive switching of the MoTe$_2$ array device after an electroforming process
(a) The electroforming process of the MoTe$_2$ array device; (b) 20 cycles I-V curves of the MoTe$_2$ array device with a compliance current of 5 mA

**2.5 Consecutive conductance modulation and neural network simulation of the MoTe$_2$ device**

Continuous pulse stimulations are applied on the device to mimic the long-term potentiation (LTP) and depression (LTD) of synapses, which are essential synaptic functions for neuromorphic computing. The conductance triggered by alternating 50 positive (1 V, 500 ns) and 50 negative (−1 V, 5 μs) input pulses exhibits repeatable and stable response of LTP and LTD of biological synapses (Fig. S11).

Fig. 6 presents one cycle of LTP and LTD realized in...
the present device. The LTP and LTD processes can be fitted by the equation below:\(^{(21-22)}\):

\[
G = a + c \times e^{-\beta N}
\]  

(1-1)

where \(G\) is conductance, \(N\) is the number of pulses, \(a\), \(c\) and \(\beta\) are constants. Here, the exponential factor \(\beta\) can reflect the linearity of conductance modulation. The smaller the \(\beta\), the better the linearity. As shown in Fig. 6, the \(\beta\) for LTP is 0.143 and the \(\beta\) for LTD is 0.203, which is comparable to the previously reported work\(^{(21)}\).

With the present device working as synapses, a fully connected network with one hidden layer has been simulated using the CrossSim platform\(^{(23)}\). The neural network is with 784 input neurons, 300 hidden neurons, and 10 output neurons for 28x28 input pixels and 10 output classifications for recognition handwritten digits from Modified National Institute of Standards and Technology (MNIST) dataset. The MoTe\(_2\) devices were used as storage of the weight in the network and the change of the conductance of the artificial synapses was adopted as the weight update in the process of the backpropagation algorithm. In addition, another network with a size of 64x36x10 was trained using another dataset of small digits with 8x8 pixels from the “Optical Recognition of Handwritten Digits” dataset. The test recognition accuracies of the simulation with two datasets using the present device are benchmarked with ideal floating-point numeric precision that represents the neuromorphic algorithm limit, as shown in Fig. 7. The recognition accuracies of the small and large digits can reach \(\approx 91.3\%\) and \(88.2\%\) after 40 epochs, lower than the ideal numeric accuracies of \(\approx 96.7\%\) and \(98.1\%\). This accuracy difference between the experiment derived and the ideal numeric results from the nonlinear and asymmetrical conductance change in LTP and LTD processes, which can be further improved through interface engineering of the device.

### 2.6 Mechanism analysis

The electron transport processes at LRS and HRS were analyzed to explain the underlying mechanism of resistive switching behavior. As shown in Fig. 8(a), nonlinear \(I-V\) characteristics are observed at the HRS, with the current increasing as the temperature increases. And it was found that the HRS was best-fitted by the Schottky emission model\(^{(24)}\) as shown in the inset of Fig. 8(a):

\[
J = A' T^2 \exp\left[-\frac{q(\phi_B - \sqrt{qE/4\pi\varepsilon_0\varepsilon_r})}{kT}\right], \quad A' = \frac{120 m^2}{m_0 k_B T^2}
\]

(1)

where \(A'\) is the effective Richardson constant, \(m_0\) is the free electron mass, \(T\) is the absolute temperature, \(q\) is the electron charge, \(\phi_B\) is the Schottky barrier height, \(E\) is the electric field across the dielectric, \(k\) is Boltzmann’s constant, \(\varepsilon_0\) is the permittivity in vacuum, and \(\varepsilon_r\) is the optical dielectric constant.

In contrast, the LRS performs Ohmic conduction due to the linear characteristic of the \(I-V\) curve (Fig. 8(b)), indicating the formation/rupture of conductive filaments might be the dominating mechanism of the resistive switching. Furthermore, the current of the LRS decreases as the temperature increases, implying the metallic filaments are formed in the LRS. These metallic filaments might be composed of the monoclinic (1T') phase (1T'-MoTe\(_2\)) resulting from phase transition of MoTe\(_2\)\(^{(25)}\) or metallic Ti filaments induced by Ti ions drifting into native defects in MoTe\(_2\)\(^{(26)}\). The detailed atomic structure of these filaments need further investigations.
3 Conclusions

Through CVD growth of the MoTe$_2$ film, the PMMA transfer method and lift-off process, the memristive devices based on 2D MoTe$_2$ film were successfully prepared. The devices exhibit stable bipolar resistive switching with superior retention characteristics and good endurance. More importantly, the devices perform high yield, low cycle-to-cycle variability, and low device-to-device variability. Furthermore, a 3x3 memristor array based on the MoTe$_2$ device with 1R scheme was successfully demonstrated. And, the simulation of neural network with the prepared MoTe$_2$ devices as synapses was implemented for handwritten digits recognition, and the recognition accuracy of around 90% was realized. Our demonstration of memristor array based on centimeter-scale 2D MoTe$_2$ film provides an avenue for future neuromorphic circuits using large-scale 2D TMDs film.

Supporting Materials

Supporting materials related to this article can be found at https://doi.org/10.15541/jim210658.

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高均一性二维碲化钼忆阻器阵列及其神经形态计算应用

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摘要：二维过渡金属硫化合物是构建纳米电子器件的理想材料，基于该材料体系开发者信息存储和神经形态计算的忆阻器，受到了学术界的广泛关注。受制于低成品率和低均一性问题，二维过渡金属硫化合物忆阻器阵列鲜见报道。本研究首次实现了基于厘米级二维 MoTe2 的忆阻器阵列，采用化学气相沉积得到厘米级二维碲化钼薄膜，并通过湿法转移和剥离工艺制备得到碲化钼忆阻器件。该碲化钼器件表现出优异的保持性（保持时间>1000 s）、快速的阻变（SET 时间~60 ns，RESET 时间~280 ns）和较好的循环寿命（阻变 2000 循环后仍可正常工作）。该器件具有高成品率（96%）、高阻变循环间差异性（SET 过程为 6.6%，RESET 过程为 5.2%）和低器件间差异性（SET 过程为 19.9%，

关键词: 二 维 材 料; 碲 化 钼; 忆阻器阵列; 神经形态计算

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1 The PMMA transfer method
2 The XRD analysis of the MoTe₂ film
3 The fabrication process of the Au/Ti/MoTe₂/Au/Ti device
4 The AFM image of the 3x3 memristor array
5 The electroforming process of the device

Before achieving a stable resistive switching, an electroforming process was required, as shown in Fig. S5. At the beginning, the current of the CVD-MoTe₂ device was low due to its high resistance. After the application of a relatively large positive voltage (about 1.15 V), the device switched to the LRS.

6 I-V curves of 24 Au/Ti/MoTe₂/Au/Ti devices
7 Cumulative distribution of device HRS and LRS of 24 devices
8 Estimation of the array size

In case of memristor crossbar array, the cross talk between the adjacent memory cells restricts the maximum possible size of the array. Especially when all the memory cells in the array are in low resistance state, the sneak path leakage problem will be highly predominant in the array. Therefore, to obtain the maximum possible crossbar array size, the worst case read scheme is utilized to measure the number of possible word lines with read margin of more than 10%, which is called the one-bit pull-up scheme.[1-3]

In this model, the bit line of the selected cell is biased to the read voltage, the word line of the selected cell is grounded and all the other word and bit lines are left floated. Regarding an N×N square crossbar array with the most challenging data pattern (i.e., all unselected cells in LRS) and negligible line resistance, the crossbar array can be simplified into three regions as shown in the left panel of Fig. S8(a). And the corresponding equivalent circuit is present in the right panel of Fig. S8(a). The resistance value of the selected cell will be found by measuring the output voltage across the pull up resistor, $R_{pu}$. To obtain the best result during the measurement, the pull up resistor value will be set to the resistance during the low resistance state ($R_{pu}=R_{LRS}$). The read margin normalized to the pull up voltage is calculated by solving the Kirchoff equation:

$$V_{out} = \frac{R_{pu}}{R_{HRS}} - \frac{R_{pu}}{R_{LRS}} \cdot \frac{V_{window}}{N-1} + R_{pu}$$

(1-1)

In order to suppress the leakage current, the memristor can be connected in series with the selector to form a 1S1R (one selector one resistor) structure. The Pt/TaOₓ/TiO₂/TaOₓ selector reported before was chosen as the selector in combination with the CVD-MoTe₂ device to form 1S1R structure since it has an I-V window that matches the CVD-MoTe₂ device (Fig. S9)[4]. The key resistance values (R_{select}^{HRS}, R_{select}^{LRS}, R_{sneak}^{LRS}) and the corresponding maximum number of word lines/bit lines (N) with read margin >10% for 1R device and 1S1R schemes are listed in Table 1. Note that $V_{read}$ is 2 V.

Fig. S8(b) shows the calculated read margin for both 1R and 1S1R schemes for different number of word lines. From this figure, it is clear seen that the read margin reduced drastically for 1R scheme and the number of word lines with at least 10% read margin is found to be 4 only. In case of 1S1R scheme, the non-linearity of the device resulted in increased number of word lines to 870 for 10% read margin, making a 740 kb possible crossbar array fabrication with good working possibility. Enhancing the selectivity of the selector can result in further increase of the crossbar array size to get a high density and large size crossbar array.

9 Memristive behavior of the array devices
10 Consecutive conductance modulation of the device

Table 1 The key resistance values and the corresponding maximum number of word lines/bit lines (N) with read margin >10%

| Scheme   | R_{select}^{HRS}/Ω | R_{select}^{LRS}/Ω | R_{sneak}^{LRS}/Ω | N    |
|----------|---------------------|---------------------|--------------------|------|
| 1R       | 150                 | 1500                | 150                | 4    |
| 1S1R     | 350                 | 1700                | 1.25×10⁵           | 870  |
Fig. S1 The PMMA transfer method

Fig. S2 XRD pattern of the MoTe$_2$ film

Fig. S3 Fabrication process of the Au/Ti/MoTe$_2$/Au/Ti device

Fig. S4 The AFM image (a) of 3×3 memristor array, and the height profile (b) along the horizontal line and the vertical line
Fig. S5 The electroforming process of the device

Fig. S6 I-V curves of 24 Au/Ti/MoTe₂/Au/Ti devices

All the devices show stable resistive switching with low cycle-to-cycle and device-to-device variability.
Fig. S7 Cumulative distribution of device (a) HRS and (b) LRS of 24 devices.

Fig. S8 Estimation of the array size for the prepared CVD-MoTe$_2$ device.
(a) Sneak current path at read in a square crossbar array where all bits except the selected one are at LRS, and the equivalent circuit can be represented by three resistors (region 1, region 2 and region 3); (b) Dependence of the read margin on the crossbar line number for both 1R and 1T1R schemes.

Fig. S9 I-V characteristics of the Pt/TaO$_x$/TiO$_2$/TaO$_x$/Pt selector.
Fig. S10 I-V curves of 9 devices in the 3x3 array

Fig. S11 Long-term potentiation and depression of the device using 50 potentiation (1 V, 500 ns) and depression (-1 V, 5 μs) presynaptic pulses.

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