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Enhancing the Matrix Addressing of Flexible Sensory Arrays by Highly Nonlinear Threshold Switch

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Abstract

The increasing need for smart systems in healthcare, wearable, and soft robotics is creating demand for low-power sensory circuits that can detect pressure, temperature, strain, and other local variables. Among the most critical requirements, the matrix circuitry to address the individual sensor device must be sensitive, immune to disturbances, and flexible within a high density sensory array. Here, we report a strategy to enhance the matrix addressing of a fully integrated flexible sensory array with an improvement of 10^8 folds in the maximum readout value of impedance by a bidirectional threshold switch. The threshold switch shows high flexibility (bendable to a radius of about 1 mm) and a high nonlinearity of $\sim 10^{10}$ by using a nano-contact structure strategy, which was revealed and validated by the molecular dynamics simulation and experiment at variable mechanical stress. Such a flexible electronic switch enables a new generation of large-scale flexible and stretchable electronic and optoelectronic systems.

Flexible electronic circuits for pressure, temperature, strain, electrochemical, and gas sensing have been recently proposed with promising applications in healthcare,^[1, 2] wearable electronics^[2-4] and robotics.^[5, 6] For such applications, the crossbar array configuration is one potential solution to enable the sensory devices to be high density, large area and mechanical flexibility.^[1, 5] However, the integration of these crossbar arrays is hindered by the notorious issue of crosstalk, which arises due to the sneak path currents from neighboring elements deteriorating the operational attributes of the array.^[5, 7] To enable the matrix addressing of the array correctly, an electronic switch possessing a nonlinear response, most typically a field-effect transistor, is usually connected to a functional element to suppress the sneak path currents.^[5, 8, 9] Recently, selection elements based on field-induced threshold switching (TS) phenomena have received considerable attention due to its two-terminal structure, low operation voltage and 3D stacking capability in crossbar arrays for high density memory circuits.^[10-12] These properties make the TS device extremely attractive for flexible electronic and optoelectronic integration, such as electronic displays, sensory circuits, information storage, and data processing. However, most of the reported TS devices are rigid and not suitable for sensor applications in healthcare, wearable and soft robotics, where high mechanical flexibility is required.

To enhance the matrix addressing of flexible sensory arrays, selection elements with high ON-state current and high nonlinearity, i.e., the ratio between the current at the read voltage (V_D) and at half read voltage ($\frac{1}{2}V_D$), are also needed.^[10, 13-15] These properties can be provided by the TS phenomena, where a conductive filament is formed in correspondence to a characteristic electric field, and dissolved spontaneously as the external electrical field is reduced below the threshold.^[10, 16] This process is controlled by two critical parameters, namely the diffusivity of migrating species and external driving forces, e.g., the surface energy (Gibbs–Thomson effect)^[11, 16-18] and mechanical stress^[16, 19] of the conductive filament.

The diffusivity of migrating species can be improved by engineering the materials, e.g., by doping the dielectric layer with fast diffusive particles.^[11, 12] Alternatively, low compliance current (CC) is another method^[20, 21] commonly used to induce the TS phenomena because it leads to the formation of thin conductive filaments with high surface energy that rupture easily and induce the OFF-state, as observed by *in situ* high resolution transmission electron microscopy.^[10, 11, 20] However, at low CC, the ON-state current level and nonlinearity may deteriorate significantly. On the other hand, the reliability of TS devices is compromised at high CC. Therefore, there is a trade-off between achieving high nonlinearity and high reliability in TS devices.

To address the current challenges of the matrix addressing of flexible sensory arrays, we developed a highly nonlinear flexible TS device as the selection element of a flexible pressure sensor array for the first time. Our TS device is based on a nanocomposite thin film consisting of an elastic insulating matrix and an electrically conducting filler of silver nanowires (AgNWs), which confers flexibility and forms a nanoscale conductive filament via the nano-contact effect. The two-terminal bidirectional flexible TS device shows an unprecedented nonlinearity of 10^{10} and good mechanical flexibility with bending radius r approaching 1 mm. To quantitatively explain the TS behavior, we proposed a switching model based on minimization of interfacial energy and thermal diffusion, and validated the model experimentally at variable mechanical stress. Finally, a fully integrated flexible pressure sensor array was experimentally demonstrated by using the highly nonlinear TS device as the selection element. The maximum readout value of impedance of a selected pressure sensor element was significantly improved with 10^8 folds in the flexible integrated pressure sensor array.

To understand the underlying mechanism of nano-contact active electrodes, we fabricated and characterized three TS devices with different contact morphologies (**Figure 1a**). Device I had a thin film silver electrode, device II and III, which contain AgNW structures in the

electrodes, took on a single-sided nanowire (SSNW) and a double-sided nanowire (DSNW) electrode configuration, respectively (see Figure S1 and Supporting Information for device configurations and fabrication process flows). To validate the universality of the nano-contact strategy, we used both silk fibroin and polydimethylsiloxane (PDMS) elastomer as the device dielectrics. Based on the evolution of current-voltage (I - V) characteristics as a function of the CC, the maximum allowed CC for reliable TS operation for devices I, II, and III are 10^{-7} , 10^{-4} , and 10^{-3} A, respectively (Figure S2). We conducted the statistical probability of TS, defined by a retention time of the ON-state resistance being less than 5 s, as a function of the CC for the three device configurations (**Figure 1b**). Compared to device I with thin film electrodes, device II and III with AgNW electrodes showed a more reliable TS behavior under high CC, and this is attributed to the nano-contact effect of the AgNW electrodes. The TS probability of $\sim 98\%$ in device III could be achieved for a broad range of CC reaching up to about 1 mA. Even for a CC of about 3 mA, the TS behavior could still be observed in device III (Figure S3a). A high CC allowed for the reliable TS behavior guarantees high nonlinearity and ON-state current of the selection operation. Note that only device III shows the bidirectional switch behavior (Figure S3b), which can be attributed to its symmetrical physical structure. These results demonstrate that the nano-contact approach can be used to improve the TS behavior, allowing highly reliable and nonlinear TS devices to be designed. The DSNW-type sample can be easily achieved by natural nano-contacts between AgNW fillers in the nanocomposite thin film, where the nano-gap can be properly mediated by the concentration of the filler.

We used the nano-contact strategy to obtain highly nonlinear flexible TS devices. The flexible TS device consists of two planar Au electrodes sandwiching an elastic composite thin film, made up of AgNWs dispersed in PDMS elastomer (**Figure 1c**; also see Figure S4 and Experimental Section for detailed fabrication process). Scanning electron microscopy images show a randomly oriented network of AgNWs embedded in the surface layer of the PDMS

elastomer (Figure S5a and b). Only a fraction of AgNWs is exposed at the very top of the PDMS elastomer, while the majority of them are firmly buried in the bulk of the elastomer, thus guaranteeing good mechanical stability of the nanostructure (inset of **Figure 1e**).^[22]

The as-prepared Au/AgNWs-PDMS/Au flexible TS device with high initial OFF resistance state ($10^{13} \Omega$) requires an electroforming process with the application of a forming voltage around 2.5 V to activate the bidirectional TS behavior (Figure S5c and d).^[10] **Figure 1d** shows the typical TS characteristics of the flexible TS device with a CC of 500 μ A after the forming process. The current is initially very low in the OFF-state until it abruptly increases to the ON-state at a threshold voltage of about 0.4 V, also referred to as the turn-on voltage, V_{ON} . The flexible TS device then spontaneously returns to the OFF-state as the applied voltage sweeps back to zero, contrary to the resistive switching behavior in memory switching devices.^[20, 21] Similar *I-V* characteristics of TS were observed under negative polarity, indicating a symmetric bidirectional switching behavior, which is highly desired for selection elements in combination with bipolar resistive switching devices.^[10, 13, 23] The maximum value of nonlinearity, which is a key figure of merit to assess the performance of a selection element, is more than 10^{10} at $V_D = 0.5$ V (**Figure 1e**), which is the highest reported in any type of flexible switches and could compete with state-of-art silicon-based electronic switches (see also **Figure 2e**).^[10, 12-14, 19, 24-29] The huge nonlinearity is also accompanied by an extremely steep slope of below $4 \text{ mV} \cdot \text{dec}^{-1}$.

For practical application of the flexible TS, device uniformity, speed, and reliability are also important aspects. **Figure 2a** shows 200 repeated *I-V* curves, which were consecutively obtained under *DC* operation in an individual flexible TS device. The data show no remarkable deterioration, indicating excellent reliability. The device-to-device uniformity was tested by collecting data from ten individual devices (Figure S6), with all devices showing repeatable and reliable bidirectional TS behavior. It should be mentioned that the turn-on voltages of the flexible TS device in this study can be intentionally tuned by

controlling the concentration of AgNWs sprayed on the silicon wafer (Figure S7), which matches well with the various functional elements in the integration array. The measured dynamical current response characteristics of the flexible TS device corresponding to a voltage pulse are shown in **Figure 2b**. The device shows fast switching (~ 520 ns; see inset of **Figure 2b**) to the ON-state under an applied pulse, but requires a relaxation time to recover to its original OFF-state after the voltage is switched to zero.^[10, 11] The characteristic relaxation time roughly ranges from 50 μ s to 400 μ s (Figure S8), in agreement with previous results.^[19] The fast relaxation characteristics of the flexible TS device are sufficient for most flexible functional sensory elements, which typically show a relaxation time of 1 ms to 10 s.^[30, 31] The flexible TS device can withstand an endurance of over 10^6 cycles under the 100 μ s/2 V programming pulses and 100 μ s/0.3 V read pulses, with a 1.8 ms wait time between each program and read pulse (Figure S9). In addition, the read disturb test showed no resistance degradation under various constant voltage stress of $V_D = \pm 0.6$ V and $1/2V_D = \pm 0.3$ V, which further demonstrates the excellent immunity to read disturbance (Figure S10).

Our flexible TS device also showed excellent mechanical stability under bending. The experimental setup for the mechanical bending test is illustrated in the Experimental Section and Figure S11a. Compared to the original state of the flexible TS device, the I - V characteristics show no obvious change under the curved state with a bending radius r of 4.9 mm (**Figure 2c**). In particular, the super nonlinear TS behavior is still observed even under extreme stress conditions of rolling on a needle of radius ~ 1 mm, the only effect being an increase of the turn-on voltage (Figure S11b), thus indicating a robust mechanical reliability. We further tested the TS behavior of the flexible TS device under both bending directions. **Figure 2d** shows the distributions of both positive (+ V_{ON}) and negative (- V_{ON}) turn-on voltages under both inward bending ($r < 0$) and outward bending ($r > 0$), as a function of the bending radius r and strain ε . The strain ε can be calculated from the r and the thickness t of the device ($t = 300$ μ m in the flexible TS device).^[32] The flexible TS device shows an

excellent mechanical bending flexibility in the strain stress of $-6\% < \varepsilon < 4\%$ with a slight change in the turn-on voltages. The turn-on voltages increase when the bending radius, r , is less than 4 mm, but the device could still maintain a typical TS behavior without any loss of ON/OFF ratio (Figure S11c), which shows the highest nonlinearity and flexibility in the reported flexible TS devices (**Figure 2e**).^[27-29] The increase of the turn-on voltages is attributed to the mechanical strain increasing the distance between two adjacent AgNWs in the PDMS elastomer (Figure S12a and b). Moreover, we tested the reversibility of flexible TS device under bending conditions ($r = 5$ mm) (Experimental Section), as shown in **Figure 2f**. The I - V characteristics of the flexible TS device during consecutive 1000 mechanical bending cycles show no noticeable deterioration. These results indicate that the flexible TS device exhibits robust mechanical stability and reliability, even under harsh substrate physical deformation, such as the twisted state (Figure S11d).

Understanding the underlying switching mechanism is vital to improve the design and performance of the flexible TS device. To probe the role of AgNWs in highly nonlinear TS behavior, we compared the behavior of the TS device with another device fabricated in exactly the same way but using a carbon nanotubes-PDMS (CNTs-PDMS) nanocomposite thin film instead. The I - V curves of the Au/CNTs-PDMS/Au devices show no reversible resistive switching behavior regardless of the concentration of CNTs (Figure S13). Compared to inert CNTs, AgNWs show high surface chemical activity and electrochemical mobility,^[16, 33] which are critical for inducing the TS behavior. Furthermore, it is well known that silver (Ag) conductive filaments are easily formed in conductive bridging random access memory (CBRAM) with an active Ag electrode.^[34, 35] Therefore, field-induced formation of Ag conductive filaments bridging adjacent AgNW clusters could explain the TS from OFF-state to ON-state. In the flexible TS device, two adjacent AgNWs might serve as the effective top and bottom electrodes to extend the percolation network in the AgNWs-PDMS nanocomposite thin film, which further identifies the Au/AgNWs-PDMS/Au device as a

DSNW-type structure (inset of **Figure 1c**).

To gain a deeper insight into the TS behavior of flexible TS device, we performed a molecular dynamics simulation based on an overdamped Langevin model (Supporting Information). In these simulations, ionic migration/diffusion is described under the influence of Joule heating, electric field, interfacial energy minimization and Gibbs–Thomson effect. The electrochemical redox reactions were neglected because their effect on the spontaneous return to the OFF-state upon removal of the external bias is negligible.^[10, 11] The simulated and experimental *I-V* characteristics of the flexible TS device under both positive and negative voltages show excellent agreement in the bidirectional TS behavior, supporting the validity of the molecular dynamics Langevin model (**Figure 3a**). The simulated dynamic evolution of the nanoscale Ag filament as a function of the sweep voltage is shown in **Figure 3b**. In the initial stage of the simulation, two silver clusters (including 30 silver nanoparticles each) are separately located nearby the two adjacent AgNWs (OFF-state in **Figure 3c**). When a positive sweep voltage is applied, the silver cluster located on the positive polarity begin to break up into nanoparticles due to localized Joule heating under electrical field, and move towards the opposite AgNW as a result of the applied electrical field and concentration gradient. Until a certain critical voltage, only a few silver nanoparticles can uniformly fill the gap between the adjacent AgNWs to form an electrically connected filament in a fast time scale (see **Figure 3b**②). Due to the CC and nanoconfinement effect of AgNWs, the formed filament should be in an extremely tiny size scale, which retains a large interfacial energy due to the Gibbs–Thomson effect.^[17] When the positive voltage decreases, the formed filament spontaneously breaks up and collapses into larger clusters to minimize the interfacial energy nearby the two adjacent AgNWs. This process is similar to Rayleigh instability of nanowires with anisotropic surface energy.^[36] Also, Nernst potential may further accelerate the dissolution of the filament. These effects eventually lead to TS behavior of the flexible TS device under both positive and negative sweep voltages. Schematic in **Figure 3c** illustrates

the volatile TS process; the formation and rupture of Ag conductive filaments bridging adjacent AgNWs results in the OFF- and ON-state of the flexible TS device.

The model also considers the role of the bending radius in the diffusive dynamics (Supporting Information). We simulated the TS behavior of the flexible TS device at the positive sweep voltage under various bending radii (**Figure 3d**), which is similar to the measured I - V curves for both inward bending ($r < 0$) and outward bending ($r > 0$). Similar simulated I - V curves were found for negative sweep voltage (Figure S12d). From the simulation results, the positive turn-on voltage ($+V_{\text{ON}}$) increases for increasing $1/r$, reaching about 3 V for $r \sim 1$ mm, in good agreement with the experimental results in **Figure 2d**. These results further demonstrate the validity of the proposed diffusive dynamics model. Note that the mechanical stress effects at the conductive filament, which were not taken into account in this model, might still have a significant role in the relaxation of the ON-state.^[19, 37]

The two-terminal transistor-free structure of the flexible TS device allowed us to develop a flexible crossbar array that integrates large-scale sensor elements for practical applications (see **Figure 4a**). In this configuration, each cross-point consists of a two-terminal flexible TS device and a sensor device connected in series to form an integrated unit (left inset of **Figure 4a**). To demonstrate this concept, we fabricated an ultrasensitive pressure sensor (see Experimental Section), consisting of a pyramidal PDMS substrate (inset of **Figure 4b**) coated by a single-walled carbon nanotubes (CNT) layer,^[30, 38] which exhibits reliable sensing characteristics as shown by the pressure-impedance relationship (**Figure 4b**). To clarify the matrix addressing mechanism from the device level, we firstly investigated the performance of an integrated unit within the crossbar array. In an integrated unit, the individual CNT pressure sensor always shows nearly linear I - V characteristics regardless of load pressure (**Figure 4c**), and the individual flexible TS device exhibits a typical TS behavior (**Figure 4d**). After integration, the current at low voltage (< 0.6 V) was effectively suppressed by the flexible TS device, while current at high bias voltage was used to accurately sense the load

pressure due to ON-state of the flexible TS device (**Figure 4e**). By setting V_D to 0.8 V, we verified that the nonlinearity of the integrated unit is as high as 10^8 for a load pressure of 748 Pa, which may effectively alleviate the crosstalk issue in sensory arrays.

Further, a fully integrated 2×2 flexible TS-based pressure sensor array was fabricated (**Figure 5a**, see Experimental Section for fabrication process). Multi-level load pressures were continuously monitored in real time for an e-skin application using the 2×2 flexible pressure sensor array. For simplicity, the flexible pressure sensor array was attached onto a 3D printed hand replica and the probe station system was used to provide and measure the real load pressure (**Figure 5b**). To evaluate the crosstalk issue, two flexible pressure sensor arrays with flexible TS-based array backplane (W/TS) and without flexible TS-based array backplane (WO/TS) were implemented to monitor the real-time and continuous load pressure. In the worst-case scenario where all unselected pixels (R_U) are subject to maximum load pressure, the selected word line (WL) is biased to V_D and the selected bit line (BL) is biased to 0 V, while all the unselected WLs and BLs are floating.^[15, 29] We can clearly see in the upper panel of **Figure 5c** that due to the leakage current flowing through the other three pixels ($R_{U1} + R_{U2} + R_{U3}$) in the WO/TS array (inset in **Figure 5c**), the impedance value of the selected pixel (R_S) was pinched to a fixed value of $2.7 \times 10^4 \Omega$ (corresponding to 1763 Pa pressure). However, sneak current path in the W/TS array was eliminated and real load pressures could be accurately detected up to $2.4 \times 10^{12} \Omega$ in the maximum readout value of impedance with a significant improvement of $\sim 10^8$ folds (lower panel of **Figure 5c**). **Figure 5d** shows the measured pressure as a function of the real load pressure of a selected pixel in two types of array; the readout range is significantly enhanced by inducing the flexible TS device into a fully integrated flexible sensory array.

To further evaluate the performance of large-scale integration array, we calculated the measurable normalized read margin at worst-case scenario in a one bit-line pull-up (OBPU) scheme (Figure S14 and Supporting Information).^[29, 39, 40] The normalized read margin versus

the number of WLs = BLs for the flexible TS-based sensory array is presented in **Figure 5e**. Here, we assume that $R_U = 10^4 \Omega$ for all unselected pixels, and $R_S/R_U = 2, 4, 8, 16, 32, 64, 128, 256, 512$ and 10^7 . The calculated maximum array size (taking 10% normalized read margin as the criterion) can reach up to 10^7 (for $R_S/R_U > 10$), which is sufficient for most electronic and optoelectronic applications in large-scale integration.

In summary, we have constructed and demonstrated a new class of flexible and super nonlinear TS device to enhance the matrix addressing of flexible sensory arrays with a significant improvement of 10^8 folds in the maximum readout value of impedance. By using the nano-contact strategy, the fabricated flexible TS device shows a super nonlinearity of 10^{10} , high ON-state current of 500 μA , and robust mechanical flexibility. The TS behavior in the nanocomposite was further revealed by molecular dynamics simulation. These results provide a promising pathway toward large-scale integration of flexible electronic and optoelectronic devices by utilizing transistor-free TS switches.

Experimental Section

Fabrication of the flexible TS device: Silver nanowires (AgNWs, 60 nm $\times$ 10 μm , 0.5 wt% in isopropyl alcohol) and polydimethylsiloxane (PDMS) were purchased from Sigma-Aldrich. Firstly, 1 mL AgNWs solution was diluted to an extremely low concentration (0.012 wt%) for inkjet printing by adding 40 mL isopropyl alcohol solution. Then, a silicon wafer after hydrophobization treatment was patterned by a shadow mask and was placed onto a hotplate of 80 $^\circ\text{C}$. Subsequently, the diluted AgNWs solution was sprayed on the silicon substrate by the inkjet printing method to form a uniform conductive film ($\sim 1 \text{ k}\Omega$) of AgNWs network. Remove the shadow mask and pour the liquid PDMS precursors with a weight ratio of 8:1 (the silicone prepolymer : the cross linker). Then, cure in an oven of 60 $^\circ\text{C}$ for 10 hours. Peel off the AgNWs-PDMS nanocomposite film from the silicon substrate. Finally, 70 nm-thickness Au planar electrodes were deposited on the region of nanocomposite film by

thermal evaporator. The space of two planar Au electrodes is 50 μm .

Fabrication of the flexible TS-based sensory array: Firstly, an ultrasensitive CNT pressure sensor array was fabricated by using a pyramidal silicon master after hydrophobization treatment. The pyramid pattern size of the silicon master was 35 μm $\times$ 35 μm . PDMS precursor was mixed in a ratio of 10:1 and was poured on the pyramidal silicon master. After curing at 60 $^{\circ}\text{C}$ for 10 hours, peel it off to obtain the PDMS substrate with pyramid patterns. Furthermore, the pyramidal PDMS substrate was patterned into a 2 $\times$ 2 array (individual cell: 1.2 mm $\times$ 1.5 mm) by the adhesive tape. 1 g L^{-1} single-walled carbon nanotube solution (in distilled water) was dispensed onto the patterned PDMS substrate after an oxygen plasma treatment with the 5 mbar, 50 W for 1 min to improve the adhesion between the PDMS and CNTs. After fast evaporating on the hotplate, remove the adhesive tape to obtain a 2 $\times$ 2 pressure sensor array. Next, a 2 $\times$ 2 flexible TS-based array backplane was fabricated by following the same process of the preparation of individual TS device, except the shadow masks with an interconnection Au layer. Finally, the CNT pressure sensor array was directly laminated on the flexible TS-based array backplane to form a fully integrated pressure sensor array.

Electrical and mechanical measurements: The current-voltage (I - V) characteristics and constant voltage stress were performed using a Keithley 4200 semiconductor device parameter analyzer. Arbitrary waveform generators (Agilent 33220A) and oscilloscope (Tektronix DPO5054B) were used to generate pulse wave and monitor the fast current response flowing through the flexible TS device. During the pulse measurement, a 1 M Ω resistor was connected in series to the flexible TS device to serve as a current limiter. The cycle testing of mechanical bending was carried out by MTS Criterion Model 42. The TS behavior of the flexible TS device was measured after it recovered to the flat state.

Supporting information

Supporting information is available from the Wiley Online Library or from the author.

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Figure 1. Nano-contact induced highly nonlinear flexible TS device. **a)** Schematic showing the configuration of TS devices with different contact morphology. I: device with a thin-film electrode, II: device with a single-sided nanowire (SSNW) electrode configuration, III: device with a double-sided nanowire (DSNW) electrode configuration. **b)** Probability of TS as a function of the CC for devices I, II, III. For simplicity of the device preparation, device I: Ag/silk/Au, device II: Ag/AgNWs/silk/Au, and device III: Ag/AgNWs-silk/Au are used to illustrate the nano-contact strategy. Data collected from five devices (20 cycles each device) for each-type device. The probability of TS under high CC was significantly improved in the nano-contact type devices (II and III), regardless of dielectrics (silk or PDMS). **c)** Schematic of the flexible TS device with two-terminal Au/AgNWs-PDMS/Au planar structure. By utilizing the natural nano-contacts of AgNWs in the nanocomposite thin film, we achieved a DSNW-type flexible TS device (inset). **d)** Typical I - V characteristics of the flexible TS

device with super nonlinear, bidirectional, and low voltage TS behavior. e) Flexible TS device with a super nonlinearity of $\sim 10^{10}$ at $V_D = 0.5$ V and a fast turn-on switching slope of < 4 mV $\cdot$ dec $^{-1}$. Inset of e) shows the optical-microscope image of the fabricated flexible TS device under twisted state.

Figure 2. Electrical and mechanical characteristics of flexible TS device. a) Consecutive 200 cycles of DC voltage sweeps with CC of 100 μ A show no deterioration. Positive and negative voltage sweeps were alternately applied. b) Dynamical response characteristics of the flexible TS device. The device can be turned on fast (520 ns; see inset) under a 10 μ s/2 V pulse, and then turned off spontaneously after removing the external electric field. The relaxation time (defined as the time required for the conductance to drop to 1/e of the maximum conductance

value after the removal of the turn-on voltage) is around 110 μs as verified by a following pulse of 480 μs /0.3 V. Here, a fixed resistor (1 M Ω) was connected in series to the flexible TS device to serve as a current limiter. c) I - V characteristics of the flexible TS device under both flat and bent state with a bending radius r of 4.9 mm. No obvious change seen under the bent state. d) Distribution of $+V_{\text{ON}}$ and $-V_{\text{ON}}$ under both inward bending ($r < 0$) and outward bending ($r > 0$) as a function of r from -2.5 mm to 1 mm. The top X-axis shows the strain ε under different bending radii. Each error bar chart was collected from 5 cycles. Good consistency between the experimental and simulated ('sim' blue line) results based on molecular dynamics model. e) Comparison of maximum nonlinearity-flexibility characteristics of the reported electronic switches with bidirectional switching behaviors. f) I - V characteristics of the flexible TS device under consecutive 1000 mechanical bending cycles show no noticeable deterioration. The bending radius r is 5 mm.

Figure 3. Simulation of molecular dynamics model. a) Simulated and experimental I - V characteristics of the flexible TS device show good agreement, validating the molecular dynamics Langevin model. CC: 0.1 mA. b) Simulated dynamic evolution of the nanoscale Ag filament between the critical adjacent AgNWs over two sweep cycles. 1st sweep cycle: applied voltage (0 V $\rightarrow$ 1.5 V $\rightarrow$ 0 V); 2nd sweep cycle: applied voltage (0 V $\rightarrow$ -1.5 V $\rightarrow$ 0 V). ①, ②, ③, ④ represent the node times when the applied voltages are 0.5 V, 1.2 V, -0.5 V, -1.2 V, respectively. With the applied voltage sweeping, Ag nanoparticles could form a conductive filament under high electrical field, and recover to their original positions as the electrical field decreases. Color bar represents the numbers of Ag atoms. In the initial state (0 V), there are totally 60 Ag atoms equally distributed within the two critical adjacent AgNWs. c) Schematic illustration of the flexible TS device switched between OFF-state and ON-state. Red cylinders represent the critical adjacent AgNWs. d) Simulated results at positive sweep voltage of the TS behaviors of flexible TS device under different bending radius are consistent

with our experimental results shown in Fig. 2d. ‘-’: inward bending ($r < 0$), ‘+’: outward bending ($r > 0$). Similar simulated result for the negative TS behaviors is in Figure S12d.

Figure 4. Integration configuration and matrix addressing mechanism. a) Schematic showing the configuration of the integrated crossbar array of sensors based on the two-terminal flexible TS devices. Left inset: Each integrated unit in the cross-point consists of a flexible TS device and a functional sensor element connected in series. Right inset: schematic of the corresponding circuit. b) Ultrasensitive pressure-impedance sensing characteristics of the CNT pressure sensor. Inset shows the SEM image of pressure sensor with pyramidal microstructures. c) Linear I - V relationship of individual pyramidal pressure sensor within an integrated unit under four different load pressures (0, 81, 120, and 748 Pa). d) Threshold I - V relationship of individual flexible TS device in the same integrated unit. e) Threshold sensing characteristics of the integrated unit under different load pressures. Current levels at low bias voltage (< 0.6 V) are suppressed by the flexible TS device. By setting $V_D = 0.8$ V, the nonlinearity of integrated unit reaches up to 10^8 for load pressure of 748 Pa.

Figure 5. Enhancing the matrix addressing in a fully integrated flexible pressure sensor array.

a) Optical image of the fabricated 2×2 flexible TS-based array backplane. b) Measurement setup of the flexible TS-based pressure sensor array. The CNT pressure sensor array was fixed on a hand replica and a probe station system was used to provide the real load pressure. Inset shows the enlarge image of a CNT pressure sensor under a load pressure state. c) Continuous sensing impedance of a selected pixel under different pressure levels in both WO/TS and W/TS array. Insets illustrate the corresponding current path in a worst-case scenario. The impedance value of the selected pixel (R_S) was fixed to $2.7 \times 10^4 \Omega$ due to the

sneak current flowing through three unselected sensors ($R_{U1} + R_{U2} + R_{U3}$). However, the undesired sneak current path is eliminated effectively in W/TS array. The maximum readout value of impedance in W/TS array was improved with $\sim 10^8$ folds. d) Measured pressure as a function of real load pressure of a selected pressure sensor in both WO/TS and W/TS array. e) Normalized read margin versus number of WLs (WLs = BLs) for a flexible integrated pressure sensor array at the worst-case scenario. Assumed that $R_U = 10^4 \Omega$ for all unselected pixels, and $R_S/R_U = 2, 4, 8, 16, 32, 64, 128, 256, 512$ and 10^7 . The calculated maximum array size can reach up to 10^7 for $R_S/R_U > 10$.

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Highly nonlinear and flexible threshold switch device was introduced to enhance the matrix addressing of flexible sensory arrays. The fabricated threshold switch shows high nonlinearity (10^{10}) and high flexibility (bendable to 1 mm) by using nano-contact structure strategy. The crosstalk issue of pressure sensor array was successfully resolved due to the high nonlinearity of threshold switch.

Nano-contact, flexible threshold switch, matrix addressing, highly nonlinear, flexible sensory array

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Enhancing the Matrix Addressing of Flexible Sensory Arrays by Highly Nonlinear Threshold Switch

ToC figure