A Light-Stimulus Flexible Synaptic Transistor Based on Ion-Gel Side-Gated Graphene for Neuromorphic Computing

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1. Introduction

With the in-depth development of the information society, the demand for big data analysis and information data processing is increasing explosively, and further requirements are put forward in terms of speed and energy consumption. The traditional von Neumann system is greatly limited by the processing speed due to the physical separation of storage and processing units, which is difficult to meet the current demands. Furthermore, it suffers from high energy consumption and large equipment volume.\textsuperscript{[1–3]} The human brain, which contains about $10^{12}$ neurons and $10^{15}$ synapses, only expends 20 W to complete complex functions such as learning, perception, reasoning, and recognition.\textsuperscript{[4,5]} Synapses serve as connections between adjacent neurons for signal transmission and exchange, and also perform learning, calculating, and remembering functions at the same time.\textsuperscript{[6]} Inspired by the powerful human brain, artificial synapses have attracted significant attention for constructing brain-like neuromorphic computing systems.

Optogenetic technology is to express the channel proteins that can be act on light for realizing the activity of neurons activated and inhibited. Here, inspired by optogenetics, a flexible side-gate artificial synaptic transistor based on ion gel and graphene is demonstrated, where the long-term remembering or forgetting could be activated by the 450 nm light stimulus. Taking advantage of in-plane-field tunable carrier transport in graphene and the high ionic motion of ion gels, various synaptic functions under flatting or bending conditions by electric stimuli are successfully mimicked, including inhibition postsynaptic current, excitatory postsynaptic current, and paired-pulse facilitation. A light-induced forgetting or remembering is demonstrated by long-term synaptic plasticity under 20 s and 10 Hz positive or negative gate pulse stimulus in the dark and 4 min recovery with light illumination. Moreover, the light stimulus promotes the associative learning and remembering function, which is demonstrated by Pavlov’s dog experiment with electrical pulses and light stimulus. Such light-assisted learning and memory function on synaptic plasticity can be attributed to the photogenerated electrons trapping effects in our device. The authors’ results provide a feasible strategy for realizing flexible light–electrical-stimulus synaptic transistors and developing the low-energy flexible neuromorphic optoelectronic hardware platform.
Synapses transmit information through the release and capture of neurotransmitters and achieve learning by changing the weights of synapses, and artificial synaptic devices are proposed and fabricated based on various materials to simulate synaptic weight changes by changing their conductance or currents. At present, artificial synapses based on electronic devices mainly include memristors\cite{7} and transistor-typed devices.\cite{12–16} Since the memristor has a single input port and its performance is greatly affected by environmental factors, it is difficult to realize a complex neural network.\cite{17} However, the use of three-terminal or multi-terminal synaptic transistors is helpful to build a more complex artificial neural computing network. Among them, synaptic transistors based on the electrical double layer (EDL) are one of the ideal candidates due to energy consumption as low as 1.23 fJ/spike.\cite{18} The EDL materials are mainly utilized in synaptic transistors such as electrolytes\cite{6,19} ion-gels\cite{16,20,21} ionic liquids\cite{22} and organic polymers\cite{23} as gate dielectrics, which can simulate the release and capture of neurotransmitters in the biology through the ion mobility and diffusivity. Usually, flexible materials, e.g., organic semiconducting polymers, carbon nanotube,\cite{24} indium zinc oxide,\cite{25} graphene,\cite{26} MoS2,\cite{27} etc. have been chosen as the channel layer materials. H. Wan et al. fabricated flexible single-wall carbon nanotube-based synaptic thin-film transistors on polyimide, and utilized thin-film ferroelectric nanogenerators (FENGs) to simulate mechanoreceptors and peripheral nerves themselves to transmit external stimulation information to synapses, which successfully simulates basic synaptic properties.\cite{28} C. Gong et al. conducted preliminary research on the short-term plasticity based on graphene/ion-gel.\cite{29} Subsequently, D. Feng et al. performed the short-term and long-term plasticity on fully flexible graphene/ion-gel-based synaptic transistors by means of circuit simulation, and demonstrated the process of learning and relearning by purely electrically simulation.\cite{30} In a word, such EDL-based synaptic transistors can be easy to manufacture on a flexible substrate for utilization in flexible brain-like neuromorphic computing circuits.

Recently, the rapid development of optogenetic technology has inspired researchers to use lights as stimulus signals\cite{31–33} Biologically speaking, optogenetic technology is that the activity of neurons can be activated or inhibited by light acting on the channel proteins.\cite{34,35} J. Jiang et al. had fabricated MoS2 photoelectric synaptic transistor gated based on biopolymer electrolyte (sodium alginate), which realized synaptic enhancement and inhibitory filtering functions from short-term synaptic plasticity (STP), and successfully simulated non-Hebb rule and Hebb rule.\cite{36} However, it does not simulate the effect of the light on the remembering and forgetting performance related to the long-term synaptic plasticity (LTP).

In this work, we propose a flexible light-stimulus artificial synaptic field-effect transistor based on graphene and ion-gel, the LTP of which is related to the remembering or forgetting is promoted or inhibited by light. Our designed synaptic transistor is fabricated on flexible substrate polyimide (PI), which has the advantages of good stability for bending state, low cost, and simplicity. Taking advantage of in-plane-field tunable carrier transport in graphene and the high ionic motion of ion-gels, various synaptic functions by electric stimuli are successfully mimicked, including inhibition postsynaptic current (IPSC), excitatory postsynaptic current (EPSC), paired-pulse facilitation (PPF), and LTP. The light can be introduced as a control signal to affect inhibition or promotion of its LTP response to an electric stimulus for simulating remembering or forgetting. The basic learning function has also been demonstrated by simulating Pavlov’s dog experiment with or without light stimulus. Our work provides an alternative strategy to affect plasticity in an optogenetic way for realizing low energy consumption and a flexible neuromorphic system.

2. Results and Discussion

Figure 1a shows that synapses, as important neuronal structures, are located between adjacent neurons. Signals are transmitted from the pre-neuron to the synapse in the form of electrical signals. When the pre-synaptic membrane senses the change of membrane potential, the synaptic vesicles on the presynaptic membrane release neurotransmitters to the synaptic cleft, and then receptors on the postsynaptic membrane receive neurotransmitters to cause the change of post-synaptic membrane potential, which leads to different response (excitatory and inhibitory response) of the postsynaptic membrane. Therefore, synapses can be divided into excitatory synapses and inhibitory synapses. Figure 1b shows the structure of our prepared device modeled on a field effect transistor. Au is sputtered on the surface of PI as a three-terminal electrode, graphene is used as a channel material to connect the drain and source electrodes, and an ion gel as a gate medium is covered on the graphene and the gate electrode. The generation of postsynaptic currents (PSCs) is one of the fundamental features of neural signaling.\cite{23} To simulate the PSC response in an artificial synaptic transistor, a series of voltage pulses are applied to the gate electrode. The anions and cations in the ion gel move under the stimulus of the gate voltage to modulate the channel carriers as mimics the release and capture of the neurotransmitters at synapses. The current between the drain and source (\(I_{DS}\)) in response to a spike is considered an action potential delivered to the postsynaptic membrane.

To characterize the electrical properties of the device, a scan of the \(I–V\) output curve is performed and its results are shown in Figure 1c. The \(V_{GS}\) changes from \(-4\) to \(4.5\) V stepped by \(0.01\) V and the bias \(V_{DS} = 0.1\) V. The curve takes on a “U” shape due to the bipolar supercurrent of the graphene. It can be seen from Figure 1c that the Dirac voltage point is around \(3\) V, that is, when the applied gate voltage is less than \(3\) V, the graphene exhibits a p-type semiconductor, and the conductive carriers are holes. Applying gate voltage over \(3\) V, the graphene is an n-type semiconductor, and the electrons become conductive carriers in graphene. By scanning the gate voltage \(-4\) V–\(4.5\) V back and forth, the Dirac point moves to \(1.2\) V and the device has an obvious hysteresis phenomenon or memristive curves, which are the key to simulating the synaptic functions. After three cycle scans, the curves basically coincide, which indicates that the device has certain robustness.

\(C–V\) and \(C–F\) tests on our device are also obtained, as shown in Figure S1, Supporting Information. Ion gel at lower gate bias (Figure S1a, Supporting Information) has a double micro-level capacitance effect to modulate channel carrier conduction, which could availably reduce the working gate voltage of the device. Under gate voltage, the abundant anions and cations in the
ion gel can mimic the movement of neurotransmitters in biological synapses. In Figure S1b, Supporting Information of the C–F curve, our fabricated device is suitable for operating at frequencies below 1 kHz, which is more fitting with the biological synapses.

The graphene in our device exhibits p-type doping characteristics, because we use a chemically transferred method to make the graphene on the PI, thus enabling a large number of defect states and the adsorption of impurities such as water molecules when exposed to air. Thus, the Fermi level of our transferred graphene film moves into the valence band, and holes become the main conductive carriers.

Figure 2 illustrates the working mechanism of our devices. Initially, without volotage, anions and cations are randomly distributed in the ion gel, and the Fermi level of graphene has no change (Figure 2b). As shown in Figure 2a, when a positive gate voltage is applied to the electrodes, the anions TFSI⁻ will move toward the gate electrode interface, while the cations EMIM⁺ will move toward the graphene surface, leading to cations and anions accumulation. Such an accumulation would form two ultra-thin capacitors, which is called EDL capacitor. Meanwhile, the Fermi level of graphene would move up but remain in the valence band, thus resulting in a reduced channel current. On the contrary, as shown in Figure 2c, a negative gate voltage would drive the cations EMIM⁺ and the anions TFSI⁻ with opposite charges, and then the Fermi level of graphene would move down, which leads to an increased channel current. Usually, in the recovery process, the diffusivity of ions is much lower than that of carriers in the graphene, so the recovery of interface states requires a relaxation time, which provides the possibility to simulate the plasticity of biological synapses.

As mentioned earlier, PSC response is a successful simulation of synaptic signaling, and changes in synaptic weights are the basis of synaptic learning and remembering processes. In such synaptic devices, changes in PSCs represent changes in salient weights, so the synaptic weight changes have been analyzed by
studying PSCs responses under different conditions of gate voltage pulses ($V_g$). PSCs can be divided into IPSC and EPSC according to the response. IPSC corresponds to the inhibition of post-synaptic membrane action potentials in biological synapses, which will lead to the inhibitory behavior of neuronal cells. On the contrary, EPSC corresponds to the excitation of post-synaptic membrane action potentials in biological synapses, which could lead to the excitatory behavior of neuron cells.

**Figure 3a** shows the change of the IPSC response to the spike stimulation pulses with different pulse widths. The IPSC response changes from 34 to 88 $\mu$A with the pulse width increasing from 25 to 150 ms. This is because, with the increase of the pulse width, more ions are accumulating at the interface and even electrochemical doping. **Figure 3b** shows the change of the IPSC response to different spike stimulation pulse voltage amplitudes. It can be seen that with the increase of the pulse voltage (from 0.5 to 2 V), the IPSC response changes from 18 to 77 $\mu$A, indicating that higher voltage can also promote ion accumulation and electrochemical doping at the interface. Therefore, the PSCs are greatly influenced by the amplitude ($V_g$) and duration ($t$) of the presynaptic pulse.

PPF is an important manifestation of short-term plasticity, which has significance in the analysis of storage and calculation in nerve synapses. PPF shows the second spike response is affected by the previous spike response, that is, when the gate is stimulated by two consecutive pulses, the channel current

![Figure 3](image-url)

**Figure 3.** a) Inhibition postsynaptic current (IPSC) versus gate pulses of same pulse amplitude (2 V) but different pulse widths. b) IPSC versus gate pulses of same pulse widths (75 ms) but different pulse amplitudes. c) The excitatory postsynaptic currents (EPSCs) triggered by a pair of presynaptic spikes ($V_g = -2 V$, $V_{DS} = 0.1 V$, $\Delta t = 25 ms$). d) Paired-pulse facilitation (PPF) index as a function of interspike interval ($\Delta t$) varying from 10 to 600 ms. The paired pulses have voltages of $-2.0 V$ and pulse width of 75 ms. e) The IPSCs triggered by a pair of presynaptic spikes ($V_g = 2 V$, $V_{DS} = 0.1 V$, $\Delta t = 25 ms$). f) PPF index as a function of interspike interval ($\Delta t$) varying from 10 to 700 ms. The paired pulses have voltages of $2.0 V$ and a pulse width of 75 ms.
formed by the latter is affected by the former, as shown in Figure 3c. The EPSC responses are triggered by two adjacent voltage spikes ($V_g = -2$ V, $\Delta t = 25$ ms, the pulse width is 75 ms). We define the EPSC peak triggered by the first pulse as $A_1$ and another EPSC peak triggered by the second pulse as $A_2$. $A_2/A_1 \times 100\%$ is called the PPF index. The PPF index has a negative exponential relationship with the pulse time interval $\Delta t$. Its function expression is as follows:[38]

$$\text{PPF index} = C_0 + C_1 \exp\left(-\frac{\Delta t}{\tau_1}\right) + C_2 \exp\left(-\frac{\Delta t}{\tau_2}\right)$$  

(1)

where $C_0$ is a constant, $C_1$ and $C_2$ are the initial facilitation degrees of the two phases, and $\tau_1$ and $\tau_2$, respectively, represent the characteristic relaxation time of the fast and slow phases. The enhanced degree of the PPF index is related to the time interval between pulses, as shown in Figure S2, Supporting Information. The PPF index test data and fitting curve ($V_g = -2$ V, $V_{DS} = 0.1$ V, $\Delta t$ varying from 10 ms to 600 ms) are shown in Figure 3d, where the PPF index is 127% at $\Delta t = 10$ ms, $\tau_1$ and $\tau_2$ are 51 and 158 ms, $C_1$ and $C_2$ are 4.86 and 24.71, respectively. $\tau_2$ is an order of magnitude larger than $\tau_1$, which is comparable to the time scale in biological synapses. As shown in Figure 3f, the IPSC responses are triggered by two adjacent voltage spikes ($V_g = 2$ V, $\Delta t = 25$ ms), and the PPF index test data and fitting curve ($V_g = 2$ V, $V_{DS} = 0.1$ V, $\Delta t$ varying from 10 to 700 ms) are shown in Figure S3, Supporting Information and Figure 3e, where the PPF index is 141% at $\Delta t = 10$ ms, $\tau_1$ and $\tau_2$ are 2.65 and 168.3 ms, $C_1$ and $C_2$ are 100.75 and 42.97, respectively. From the aforementioned test results, when $\Delta t$ is less than the ion relaxation time, the ions triggered by the previous pulse cannot return to the original state before the next pulse, so a PPF phenomenon occurs. The fewer residual ions at the ion-gel interface, the lower the PPF index is. Furthermore, we speculate that the time constants $\tau_1$ and $\tau_2$, respectively, corresponding to the fast and slow phases are related to the ionic mobility and diffusivity and relaxation state of the graphene/ion-gel interface.

To prove the flexibility and stability of the device, we investigate the PPF characteristic test when the device is bent at about 60°, and the test results are shown in Figure 4. Under the bending state, the PPF index under negative gate voltage is shown in Figure 4b, where $\tau_1$ and $\tau_2$ are 5.7 and 84.1 ms, and the PPF index is 124.3% at $\Delta t = 10$ ms (Figure 4a and Figure S4, Supporting Information), which is close to the PPF index of the same stimulation pulse in the natural state (Figure 3c). Figure 4d shows that the PPF index curve under positive gate voltage when the device is bent about 60°, and $\tau_1$ and $\tau_2$ are 4.88 ms and 105.3 ms, and the PPF index is 139% at $\Delta t = 10$ ms (Figure 4c and Figure S5, Supporting Information), which is also close to the PPF index of the same stimulation pulse in the natural state (Figure 3e).

We mimic the IPSC responses with multiple presynaptic electrical pulses stimulus under 2 V gate voltage and different frequencies. Figure 5a shows the IPSC responses with different frequency pulse trains (1, 5, 10, and 20 Hz), where the pulse width and the time are fixed at 50 ms and 20 s, respectively.

![Figure 4. PPF characteristic test when the device is bent about 60°.](image-url)

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Adapted from: [51x660]A as 75 ms). We define the EPSC peak triggered by the first pulse as $A_1$ and another EPSC peak triggered by the second pulse as $A_2$. $A_2/A_1 \times 100\%$ is called the PPF index. The PPF index has a negative exponential relationship with the pulse time interval $\Delta t$. Its function expression is as follows:[38]

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We mimic the IPSC responses with multiple presynaptic electrical pulses stimulus under 2 V gate voltage and different frequencies. Figure 5a shows the IPSC responses with different frequency pulse trains (1, 5, 10, and 20 Hz), where the pulse width and the time are fixed at 50 ms and 20 s, respectively.
The IPSC amplitude gain is defined as $A_{20}/A_1$, where $A_{20}$ and $A_1$ respectively, correspond to the amplitude of IPSC after the 20th and 1st pulses, IPSC responses versus pulse frequency is plotted in Figure 5b. It can be clearly seen that, with the increase of the pulse frequency, the IPSC amplitude gain related to synaptic weight increases monotonically from 116.7% to 354.4%, which proves that our device could realize the transition from STP to LTP by improving the pulse frequency. The reason for the phenomenon is that the time interval between pulses is shortened with the increase of the pulse frequency, thus decreasing the probability of ions involved in diffusion and relaxation, thereby increasing the ions accumulation, and then resulting in an increase of the IPSC amplitude gain.

As we all know, the activity of neurons can be activated or inhibited by light. Here, the activation or inhibition of synaptic devices by light stimulus based on the principle of optogenetics has been simulated. Figure 6a shows a schematic diagram of blue light illumination on the device, where the light (30 mW, $\lambda = 450$ nm) illuminates the graphene channel. The photogenerated electrons trapping process at the graphene/ion-gel interface is schematically shown in Figure 6b. Under light illumination (30 mW, $\lambda = 450$ nm), the graphene channel generates photogenerated carrier pairs. Due to the existence of trap centers, e.g., inherent defects, hanging bonds, and local structural distortion, photogenerated electrons could be partially trapped on the surface, while photogenerated holes as free carriers could participate in the channel conduction for increasing the channel current, thereby achieving the inhibition or promotion of electrically stimulated LTP effects related with remembering and forgetting.

Figure 6c shows the LTP effect is stimulated by the positive gate voltage pulse sequence with a duration of 20 s, frequency of 10 Hz, and amplitude of 2 V. The change of the IPSC from the beginning to the end of the pulse sequence in Figure 6c is, respectively, defined as $A_2$ or $C_1$, and the change of the IPSC from the end of the pulse sequence to 4 min silence after electric pulse stimulus in Figure 6c is, respectively, defined as $B_2$ or $C_2$. As shown in Figure 6d, we first apply a sequence of electrical pulses (2 V and 10 Hz gate voltage) with a duration of 20 s to stimulate the IPSC, and then utilize the 450 nm blue light with a power of 30 mW to illuminate our devices for activating or inhibiting the postsynaptic current. The $B_2$ or $C_2$ (or $C_2/C_1 \times 100\%$) is defined as the degree of recovery. It is found that the IPSC recovered 42.83% silently after 4 min without light illumination, and the IPSC recovered 61.36% after 4 min under blue light illumination. This proves that 450 nm light stimulus on the devices presents a promotion recovery on the LTP effect related to electric stimulus under positive gate-voltage pulses, which corresponds to the forgetting property of the synapse.

Figure 6e,f shows the LTP effects of the device, which is stimulated by negative gate-voltage pulses (~2 V and 10 Hz) without and with light illumination, respectively. In Figure 6e,f, the $D_2/E_1$ (or $E_2/E_1 \times 100\%$) is defined as the degree of recovery. After calculation, it is found that the EPSC recovered 47.5% silently after 280 s (Figure 6e), and the EPSC recovered 38.7% after 280 s under the blue (450 nm) light illumination (Figure 6f), which proves that light has an inhibition recovery on the LTP effect corresponding to the remembering property of the synapse. These results demonstrate that light can affect the LTP under different voltage electric stimuli, either promoting or inhibiting it.

Associative learning as one of the simplest learning forms, also known as Pavlov’s dog experiment, plays a key role in individual adaptability and the learning of the brain. Here, ion-carrier hysteresis and photogenerated electrons trapping effects enable our device to mimic the classical associative learning behavior. In the Pavlovian experiment, before the learning training, the fed food was called unconditioned stimulus (US), and unconditioned response (UR), respectively. The dog salivates to the food (US) but not to the bell (NS). After repeated training, bells established an association with food, at which point NS changed to the conditional stimulus (CS). In our device, the time-dependent LTP related with or without light stimulus is the key plasticity in mimicking associative learning.
Figure 7a, the pulses (5 Hz) with the amplitude of $-0.5$ and $-3$ V are, respectively, defined as NS and US signals. It can be seen in Figure 7a that before applying and training progress, 10 s NS and 10 s US signals are implemented on the presynaptic terminal separately. An output current of $420 \mu A$ is set as the threshold for the "salivation" response. Before training progress, in Figure 7b, the EPSC related to NS signals alone is less than the threshold, that is, it is in a dog-free state, and the EPSC with US signals alone stimulus reaches the threshold, that is, it is in a dog-salivation state. Subsequently, a $-3$ V pulse (US) followed by a $-0.5$ V pulse (NS) (in Figure 7a) is alternately applied to the presynaptic terminal as the training process, corresponding to the simultaneous giving of food and bell stimulus to obtain a high EPSC over the threshold. After the training and waiting 2 min, the NS signals are alone applied as the stimulus, where a response current close to the threshold is observed in Figure 7b. This indicates that the NS and US training briefly is associated with learning, and the NS is changed to CS but not up to the "salivation" response level due to insufficient training intensity. After waiting 3 min, the EPSC related with a repeated $-0.5$ V pulses alone remains low to the threshold ($420 \mu A$), that is, the brain elimination of conditioned learning and forgetting (Figure 7b).

According to the principle of optogenetics, a 450 nm blue light with 30 mW has been introduced as a light stimulus after the NS and US training and learning (Figure 7c). After waiting 2 min, the EPSC related with a $-0.5$ V pulses alone remains low to the threshold ($420 \mu A$), that is, the brain elimination of conditioned learning and forgetting (Figure 7b).
and US. Subsequently, keeping the light illuminating for 3 min, the conditioned response applied with the CS signal (−0.5 V pulses with 5 Hz frequency) is still maintained, which indicates that light stimulus could be utilized to effectively improve the conditioned learning and training ability of the device and promote the long-term memory to reach the “salivation” response level. In a word, the light and electric stimulus applied in our device could realize associative learning and remembering.

3. Conclusion

In conclusion, a side-gate fully flexible light–electrical-stimulus synaptic transistor based on graphene and ion gel has been fabricated. The ions in the ion gel simulate the transmission of neurotransmitters in biological synapses, the gate acts as a presynaptic membrane to receive signals, and the neural excitation and inhibition functions are achieved by applying different gate voltage pulses and light stimulus. Basic biological functions, including IPSC, EPSC, PPF, and LTP, were successfully mimicked. More importantly, based on the principle of optogenetics, the 30 mW blue light stimulus (450 nm) has been utilized to realize the promotion or inhibition of the LTP effect of the device, due to the photo-generated electrons trapping effect. Moreover, Pavlov’s dog experiment has been simulated by electric and light stimulus, which applied in our device could realize the associative learning, forgetting, and remembering function. Therefore, the ion-carrier hysteresis and photogenerated electrons trapping effects enable our device with a light and electric stimulus to mimic the synaptic plasticity and the classical associative learning behavior. Our proposed synaptic transistors would provide new opportunities for the development of artificial synapses and their application in future flexible optoelectronic neuromorphic computing systems.

4. Experimental Section

Device Fabrication: PI was used as the substrate material, planar gate-drain-source Au electrodes were fabricated on PI by sputtering and lithography technology, and then the bilayer graphene film was transferred between the source-drain electrodes by polymethylmethacrylate wet transfer method. The prepared ionic glue was dripped on the graphene and gate electrodes, and then the volatile solvent was removed with a hot plate at 100 °C.

Characterization Measurement: I–V, C–V, and C–F curve tests were performed under the Keithley 4200 semiconductor characterization system, and the remaining electrical measurements were performed on the Keithley 2450 source meter by KickStart 2.0 software. The gate voltage pulse train is generated by an arbitrary function signal generator Siglent SDG 5162. The ADR-1805 produces blue light with a wavelength of 450 nm.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.
Keywords

artificial synapses, electric-double-layers, flexible devices, ion-gel side-gated graphene transistors, light stimulus