All-Optical Reconfigurable Electronic Memory in a Graphene/SrTiO3 Heterostructure

Liyun Qin, Qinliang Li, Shiteng Wu, Jianyu Wang, Zhendong Wang, Li Wang,* and Qisheng Wang*

Cite This: ACS Omega 2022, 7, 15841−15845

ABSTRACT: Direct optical data coding in an electronic device is meaningful for photonic technology. Herein, we report electronic memory in a graphene/SrTiO3 heterostructure, which presents the all-optical logic operation (encoding and decoding). The underlying physics have been elucidated in which the synergistic effect of surface localization with interface band bending is responsible for optical encoding and decoding in the electronic memory device of the graphene/SrTiO3 heterostructure. Further, we demonstrate a robust retention and synaptic-like processing of optical signals, which may lead to significant applications in neuromorphic imaging sensors.

INTRODUCTION

Optical data processing,1−3 featuring a high bandwidth and low power dissipation, is central to next-generation information technology such as big data, cloud computing, and artificial intelligence. Meanwhile, electrical data processing based on silicon field-effect transistors is still the dominating information operation principle in modern communication and memory technology because of high controllability of electrons and mature manufacture of integrated electrical circuits.4−6 However, so far, optical information processing is always incompatible with integrated electrical circuits. The combination of optical transmission and data processing strategies with electrical chip memory technology is promising to break through the bottleneck of the state-of-the-art information industry.

2D material-based optoelectronic random memory (ORM) has recently renewed the researchers’ interest due to its intriguing electronic properties.7,8 2D materials like graphene,9 MoS2, black phosphorus, and boron nitride with atomic-scale thin thickness strongly interact with external perturbations such as an electrical field,10−14 surface adsorbrates, and interface impurities.15,16 Together with strong light−matter interactions,17 2D material-based ORM devices present high endurance and reproducibility,8 multibit memory,15,19 and CMOS compatibility.20 However, the reported ORM devices are generally operated through optical writing and electrical erasing, which have not yet completely applied photons to control electronic memory.21−24 The all-optical manipulation of electronic memory devices will enable the integration of photonics with electronic circuits.

Herein, we propose all-optically manipulated ORM in a graphene/SrTiO3 heterostructure where the data is fully written and erased via light pulses. Optical information is stored by electrical methods, which connect the optical data manipulation with electrical information memory together. The behind mechanism has been elucidated via controlling the surface absorbates and photon energy. Lastly, the stability and multibit states have been demonstrated, indicating a neuromorphic imaging sensor application.

EXPERIMENTAL METHODS

Materials and Characterization. The all-optical manipulation of electronic memory was performed on two-terminal graphene on strontium titanate (SrTiO3). SrTiO3 with an area of 1 × 1 cm was bought from HF-Kejing Company. Monolayer graphene, which was transferred into the SrTiO3 substrate, was purchased from 6C Company. As shown in Figure S1, the composition was confirmed by Raman spectra (DXR3 laser confocal PL & Raman microscopy spectrophotometer). The excitation wavelength was 532 nm with a maximum laser power of 10 mW.

Optoelectronic Measurements. For characterizing the optoelectronic properties, the copper electrodes were fabricated using an electron beam evaporation system with a

Received: February 15, 2022
Accepted: April 14, 2022
Published: April 26, 2022
hard mask. Electrical transport was measured by a Keithley 2400 source meter at room temperature in a homemade probe station. TTL mode laser diodes with wavelengths of 405, 520, 658, 780, 850, and 980 nm were applied to irradiate the samples. A signal generator RIGOL LXI 2 was applied to adjust the laser output frequency.

RESULTS AND DISCUSSION

Photoresponse Properties. The output characteristic curves of graphene under illumination of 520 and 405 nm lasers are shown in Figure 1a,b. The laser is focused on a channel with powers of 30 and 5 mW for 520 and 405 nm lasers, respectively. The channel length is around 2 mm. Interestingly, the graphene device presents positive response at 520 nm illumination where the current increases while negative response at 405 nm illumination where the current decreases. The photoresponse (Figure S2) of 658, 780, 850, and 980 nm all shows positive response. As shown in Figure 1c, the photocurrent $I_{ph} = I_{light} - I_{dark}$ changes from $-39.5$ to $40.0$, $32.5$, $10.5$, $17.6$, and $37.1 \mu$A as the light wavelength increases from 405 to 520, 658, 780, 850, and 980 nm, respectively. The laser powers of 405, 520, 658, 780, 850, and 980 nm are 5, 30, 40, 40, 40, and 40 mW, respectively.

Reconfigurable Optoelectronic Memory. We next demonstrate the all-optical manipulation of electrical memory of graphene based on persistent positive and negative photocurrent effects. As shown in Figure 2a, a one-second 520 nm laser pulse induces a spontaneous increase in current. The $I_{off}$ and $I_{on}$ respectively represent the dark current and current with light on. Then, the current exponentially decays to a stable current value ($I_{persistent}$). The persistent positive photoresponse can also be seen with 658 and 850 nm laser illumination (Figure S3). The negative persistent photocurrent can be realized by irradiation of a one-second 405 nm laser pulse as shown in Figure 2b. The current sharply drops and comes to a stable value with the application of a 405 nm laser pulse. Therefore, an all-optical control of data writing and erasing of electrical memory can be realized as shown in Figure 2c. The continuous 520 nm laser pulses followed with 405 nm laser pulses constitute the reproducible encoding and decoding.

Figure 1. Laser wavelength-modulated photoresponse in the graphene/SrTiO$_3$ heterostructure. The 520 and 405 nm lasers respectively induce (a) positive and (b) negative photoresponse. (c) Photocurrent $I_{ph}$ changes from negative to positive as the laser wavelength increases from 405 to 520, 658, 780, 850, and 980 nm. The $I_{ph}$ is extracted at a voltage bias of 1 V.

Figure 2. All-optical reconfigurable electronic memory. (a) A 520 nm laser pulse induces the persistent photocurrent. (b) Erasing operation of a 405 nm laser pulse. The pulse duration is 1 s with powers of 30 and 5 mW for 520 and 405 nm lasers, respectively. (c) Reconfigurable electronic memory of graphene with optical encoding (520 nm laser pulses) and decoding (405 nm laser pulses).
function, respectively. The readout current is obtained under a voltage bias of 1 V in all measurements.

**Effect of Surface Absorbents.** In order to illuminate the underlying physics of persistent photocurrent effects in graphene, we first annealed the graphene device in a furnace at 400 °C with a duration of 20 min. The tube was fed with a 150 sccm inert argon atmosphere. As shown in Figure 3a, both 658 nm laser-induced positive and 405 nm laser-induced negative photoresponse disappear after annealing. The current–voltage curve with light on repeats that under dark conditions. Therefore, the surface absorbents should be the origin of persistent photoconduction effects in graphene. This would be further confirmed by the experiment of the same devices, which were exposed to ambient for 24 days. As shown in Figure 3b, the positive and negative photoresponse is completely recovered. The device again shows persistent and erasable photocurrent as shown in Figure 3c. The 405 and 520 nm lasers with a pulse width of 1 s and a frequency of 1 Hz were applied to illuminate the samples.

**Memory Mechanism and Performance.** The above results point out that the surface absorbents are central to the reconfigurable all-optical processing on electrical memory of graphene. As shown in Figure 4a, the p-type doping of graphene originates from the presence of oxygen functional groups on the surface of graphene.27,28 The Fermi level ($E_F$) resides in the valence band. According to previous theoretical calculation of electronic density of states, oxygen brings out strongly localized and half-filled states at the Fermi level.29 The photon with energy $h\nu \geq 2E_F$ allows the transition of an electron from the valence to conduction band. The laser with a wavelength of 520−980 nm excites holes and electrons.30 The electrons are captured by electron-trapping centers at the Fermi level. The localized electrons act as the negative gating effect, which accumulates holes in the channel and induces a persistent and positive photocurrent in p-type graphene. However, the short-wavelength laser (405 nm) with a large photon energy of 3.1 eV induces an electron transition from the valence band to the conduction band of SrTiO$_3$.31 As shown in Figure 4a, the energy alignment between graphene and SrTiO$_3$ helps to foster an efficient carrier injection from graphene to SrTiO$_3$. This enables the device to exhibit a strong photocurrent response to the laser illumination. The high carrier density and low carrier mobility in the channel lead to a large resistive switching ratio of the device. The device shows stable and endurable on and off states. The electron trapping effect in the graphene device enables the device to exhibit a high resistive switching ratio.27,28 The device shows low on-off ratio of 10$^{-3}$ due to the low electron mobility in the graphene channel.

Figure 3. Effect of surface absorbents on the memory effect. (a) After annealing, the positive and negative photoresponse disappears. (b,c) After exposure of the samples to ambient for 24 days, the devices recover their original states. (b) Low (wavelength, 658 nm)/high (wavelength, 405 nm)-energy photons induce the positive/negative photoresponse. (c) Low (wavelength, 520 nm)/high (wavelength, 405 nm)-energy photons arouse positive/negative persistent photoresponse.

Figure 4. Mechanism and performance of all-optical reconfigurable electronic memory. (a) Operation principle of optical writing and erasing: low-energy photon-induced electrons are localized, which induces positive and persistent photocurrent, and the photons with energy higher than that of SrTiO$_3$ excite holes, which transfer into graphene through the interface built-in field and neutralize the localized electrons. (b) Devices show stable and endurable on and off states. (c) Neuromorphic function of potentiation and depression through applying illumination of continuous 520 and 405 nm laser pulses.
and SrTiO$_3$ presents a band bending with a built-in field from SrTiO$_3$ to graphene.$^{32}$ The photoexcited holes in SrTiO$_3$ are injected to graphene. Then, the injected holes neutralize localized electrons, resulting in the reduction of current. Therefore, a 405 nm laser erases the persistent positive photocurrent. Also, graphene presents the persistent negative conductivity behavior.

We finally evaluate the performance of our all-optical operated electrical memory of graphene. As shown in Figure 4b, the memory on and off states are highly stable within 24 cycles. In addition, the multistate writing and erasing operations are realized through irradiation of 520 and 405 nm laser pulses with a 1 Hz frequency. This indicates a pulse-tunable synaptic potentiation/depression function with neuromorphic computing application in the future. However, our electric memory has a low on/off ratio due to the semimetallc nature of graphene. To address the low on/off ratio in graphene, one may use the other alternative two-dimensional materials like MoS$_2$ with semiconductive properties in the future. The writing and erasing speed of nanosecond scale is another obstacle for practical application. The slow response likely originates from the surface/interface disorders like charged surface states and impurities, substrate surface roughness, and substrate optical phonons. The response speed may be improved through packing of graphene with boron nitride or PMMA,$^{33}$ which will also improve the cyclability of the devices. Despite these, the all-optical controlled electrical memory of graphene fully utilizing the photon properties of low power dissipation would generate applications of optical data processing that does not require a large communication bandwidth.

**CONCLUSIONS**

In summary, we report an all-optically operated electronic memory device that encodes and decodes signals through light pulses. This changes the work modes of conventional optoelectronic memory in which light writes the signals while electricity erases information. The all-optical reconfigurable electronic memory of graphene originates from surface absorbates that generate the localization states of electrons. The all-optical coding of electronic memory provides a strategy for full integration of optical and electrical information technology, which may lead to fascinating applications in optical data memory and artificial visual systems.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c00938.

Raman spectra of graphene/SrTiO$_3$ and graphene/Si heterostructures and photoresponse of the graphene/SrTiO$_3$ heterostructure with irradiation of 658, 780, 850, and 980 nm lasers (PDF)

**AUTHOR INFORMATION**

Corresponding Authors

Li Wang — Department of Physics, Nanchang University, Nanchang 330031, China; orcid.org/0000-0001-6919-1712; Email: liwang@ncu.edu.cn

Qisheng Wang — Department of Physics, Nanchang University, Nanchang 330031, China; orcid.org/0000-0002-9913-7693; Email: wangqs@ncu.edu.cn

**Authors**

Liyun Qin — Department of Physics, Nanchang University, Nanchang 330031, China

Qinliang Li — Jiangxi Key Laboratory of Nanomaterials and Sensors, School of Physics, Communication and Electronics, Jiangxi Normal University, Nanchang 330022, China

Shiteng Wu — Department of Physics, Nanchang University, Nanchang 330031, China

Jianyu Wang — Department of Physics, Nanchang University, Nanchang 330031, China

Zhendong Wang — Department of Physics, Nanchang University, Nanchang 330031, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c00938

**Author Contributions**

L.Q. and Q.L. contributed equally to this work.

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

The work is supported by the National Natural Science Foundation of China under Grant Nos. 62005112, 61764011, 11727902, and 61904055. L.W. acknowledges support from the Jiangxi Provincial Innovation Talents of Science and Technology (Grant No. 20165BCB18003). Z.W. acknowledges support from the Natural Science Foundation of Jiangxi Province (Grant No. 20212B0201030).

**REFERENCES**

(1) Kuramochi, E.; Nozaki, K.; Shinya, A.; Takeda, K.; Sato, T.; Matsuo, S.; Taniyama, H.; Sumikura, H.; Notomi, M. Large-scale integration of wavelength-addressable all-optical memories on a photonic crystal chip. Nat. Photonics 2014, 8, 474–481.

(2) Nozaki, K.; Lacraz, A.; Shinya, A.; Matsuo, S.; Sato, T.; Takeda, K.; Kuramochi, E.; Notomi, M. All-optical switching for 10-Gb/s packet data by using an ultralow-power optical bistability of photonic-crystal nanocavities. Opt. Express 2015, 23, 30379–30392.

(3) Shcherbakov, M. R.; Liu, S.; Zubyuk, V. V.; Vaskin, A.; Vabisichchevich, P. P.; Keeler, G.; Pertsh, T.; Dolgova, T. V.; Staude, L.; Brener, I. Ultrafast all-optical tuning of direct-gap semiconductor metasurfaces. Nat. Commun. 2017, 8, 1–6.

(4) Ionescu, A. M.; Riel, H. Tunnel field-effect transistors as energy-efficient electronic switches. Nature 2011, 479, 329–337.

(5) Schreiber, L. R.; Bluhm, H. Toward a silicon-based quantum computer. Science 2018, 359, 393–394.

(6) Graham, A. H. D.; Robbins, J.; Bowen, C. R.; Taylor, J. Commercialisation of CMOS integrated circuit technology in multielectrode arrays for neuroscience and cell-based biosensors. Sensors 2011, 11, 4993–4997.

(7) Xiang, D.; Liu, T.; Xu, J.; Tan, J. Y.; Hu, Z.; Lei, B.; Zheng, Y.; Wu, J.; Neto, A. C.; Liu, L.; Chen, W. Two-dimensional multibit optoelectronic memory with broadband spectrum distinction. Nat. Commun. 2018, 9, 1–8.

(8) Zhou, F.; Chen, J.; Tao, X.; Wang, X.; Chai, Y. 2D materials based optoelectronic memory: conveyance of electronic memory and optical sensor. Research 2019, 2019, 1–17.

(9) Geim, A. K.; Novoselov, K. S., The rise of graphene. In Nanoscience and technology: a collection of reviews from nature journals, World Scientific 2010, 6, 11–19.

(10) Liu, F.; Zhu, C.; You, L.; Liang, S. J.; Zheng, S.; Zhou, J.; Fu, Q.; He, Y.; Zeng, Q.; Fan, H. J.; Ang, L. K.; Wang, J.; Liu, Z. 2D Black Phosphorus/SrTiO$_3$-Based Programmable Photocductive Switch. Adv. Mater. 2016, 28, 7768–7773.
(11) Ganatra, R.; Zhang, Q. Few-layer MoS2: a promising layered semiconductor. ACS Nano 2014, 8, 4074−4099.
(12) Li, L. H.; Chen, Y. Atomically thin boron nitride: unique properties and applications. Adv. Funct. Mater. 2016, 26, 2594−2608.
(13) Lei, S.; Wen, F.; Li, B.; Wang, Q.; Huang, Y.; Gong, Y.; He, Y.; Dong, P.; Bellah, J.; George, A.; Ge, L.; Lou, J.; Halas, N. J.; Vajtai, R.; Ajayan, P. M. Optoelectronic memory using two-dimensional materials. Nano Lett. 2015, 15, 259−265.
(14) Gruber, E.; Wilhelm, R. A.; Pétyua, R.; Smekal, V.; Kozubek, R.; Hierzenberger, A.; Bayer, B. C.; Aldazabal, I.; Kazansky, A. K.; Libisch, F. Ultrafast electronic response of graphene to a strong and localized electric field. Nat. Commun. 2016, 7, 1−7.
(15) Pinto, H.; Markevich, A. Electronic and electrochemical doping of graphene by surface adsorbates. Beilstein J. Nanotechnol. 2014, 5, 1842−1848.
(16) Bandurin, D. A.; Torre, I.; Kumar, R. K.; Ben Shalom, M.; Tomadin, A.; Principi, A.; Aton, G. H.; Khestanova, E.; Novoselov, K. S.; Grigorieva, I. V.; Ponornenko, L. A.; Geim, A. K.; Polini, M. Negative local resistance caused by viscous electron backflow in graphene. Science 2016, 351, 1055−1058.
(17) Li, Z.-W.; Hu, Y.-H.; Li, Y.; Fang, Z.-Y. Light−matter interaction of 2D materials: Physics and device applications. Chin. Phys. B 2017, 26, No. 036802.
(18) Lee, D.; Hwang, E.; Lee, Y.; Choi, Y.; Kim, J. S.; Lee, S.; Cho, J. H. Multibit MoS2 photoelectronic memory with ultrahigh sensitivity. Adv. Mater. 2016, 28, 9196−9202.
(19) Tran, M. D.; Kim, H.; Kim, J. S.; Doan, M. H.; Chau, T. K.; Vu, Q. A.; Kim, J. H.; Lee, Y. H. Two-terminal multibit optical memory via van der Waals heterostructure. Adv. Mater. 2019, 31, 1807075.
(20) Marega, G. M.; Zhao, Y.; Avsar, A.; Wang, Z.; Tripathi, M.; Radenovic, A.; Kis, A. Logic-in-memory based on an atomically thin semiconductor. Nature 2020, 587, 72−77.
(21) Lu, M. P.; Lu, M. Y.; Chen, L. J. Multibit Programmable Optoelectronic Nanowire Memory with Sub-femtouloule Optical Writing Energy. Adv. Funct. Mater. 2014, 24, 2967−2974.
(22) Wang, Q.; Wen, Y.; Cai, K.; Cheng, R.; Yin, L.; Zhang, Y.; Li, J.; Wang, Z.; Wang, F.; Wang, F.; Shifa, T. A.; Jiang, C.; Yang, H.; He, J. Nonvolatile infrared memory in MoS2/PhS van der Waals heterostructures. Sci. Adv. 2018, 4, eaap7916.
(23) Roy, K.; Padmanabhan, M.; Goswami, S.; Sai, T. P.; Ramalingam, G.; Raghavan, S.; Ghosh, A. Graphene−MoS2 hybrid structures for multifunctional photoresponse memory devices. Nat. Nanotechnol. 2013, 8, 826−830.
(24) Yin, L.; He, P.; Cheng, R.; Wang, F.; Wang, F.; Wang, Z.; Wen, Y.; He, J. Robust trap effect in transition metal dichalcogenides for advanced multifunctional devices. Nat. Commun. 2019, 10, 1−8.
(25) Zhou, F.; Zhou, Z.; Chen, J.; Choy, T. H.; Wang, J.; Zhang, N.; Lin, Z.; Yu, S.; Kang, J.; Wong, H.-S. P.; Chai, Y. Optoelectronic resistive random access memory for neuromorphic vision sensors. Nat. Nanotechnol. 2019, 14, 776−782.
(26) Morozov, S. V.; Novoselov, K. S.; Katsnelson, M. I.; Schedin, F.; Elias, D. C.; Jaszczak, J. A.; Geim, A. K. Giant intrinsic carrier mobilities in graphene and its bilayer. Phys. Rev. Lett. 2008, 100, No. 016602.
(27) Chen, J.; Zhu, C.; Cao, G.; Liu, H.; Bian, R.; Wang, J.; Li, C.; Chen, J.; Fu, Q.; Liu, Q.; Meng, P.; Li, W.; Liu, F.; Liu, Z. Mimicking neuromplasticity via ion migration in van der Waals layered copper indium thiophosphate. Adv. Mater. 2021, 2104676.
(28) Chen, J.-H.; Jang, C.; Adam, S.; Fuhrer, M. S.; Williams, E. D.; Ishigami, M. Charged-impurity scattering in graphene. Nat. Phys. 2008, 4, 377−381.
(29) Biswas, C.; Güneş, F.; Loc, D. D.; Lim, S. C.; Jeong, M. S.; Pribat, D.; Lee, Y. H. Negative and positive persistent photocurrent in graphene. Nano Lett. 2011, 11, 4682−4687.
(30) Bao, Q.; Loh, K. P. Graphene photonics, plasmonics, and broadband optoelectronic devices. ACS Nano 2012, 6, 3677−3694.
(31) Hasegawa, T.; Shirai, M.; Tanaka, K. Localizing nature of photo-excited states in SrTiO3. J. Lumin. 2000, 87-89, 1217−1219.