

Supporting Information

Floating-Gate Field-Effect Transistor Memory Based on Organic Crystals with Built-In Tunneling Dielectric by One-Step Growth Strategy

Zichen Chen, Shuai Chen, Tianhao Jiang, Shuang Chen, Ruofei Jia, Yanling Xiao, Jing Pan,
Jiansheng Jie, and Xiujuan Zhang**

Z. C. Chen, Prof. S. Chen, Dr. T. H. Jiang, S. Chen, R. F. Jia, Y. L. Xiao, J. Pan, Prof. J. S. Jie and Prof. X. J. Zhang

Institute of Functional Nano & Soft Materials (FUNSOM)

Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices

Soochow University

Suzhou, Jiangsu 215123, P. R. China

E-mail: xjzhang@suda.edu.cn; shuaichen@suda.edu.cn

Prof. J. S. Jie

Macao Institute of Materials Science and Engineering

Macau University of Science and Technology

Taipa, Macau, SAR 999078, P. R. China

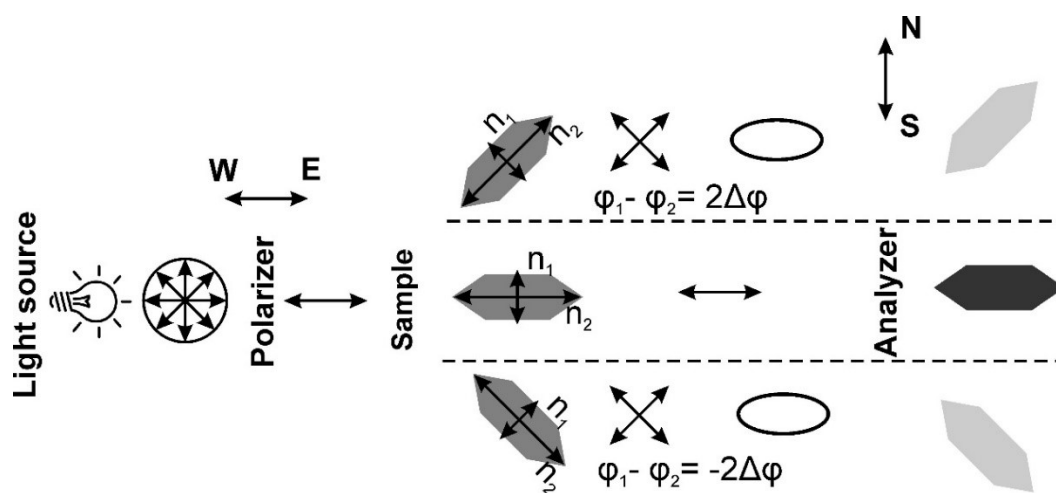


Figure S1. Schematics of light propagation in an anisotropic crystal during CPOM in reflected configuration.

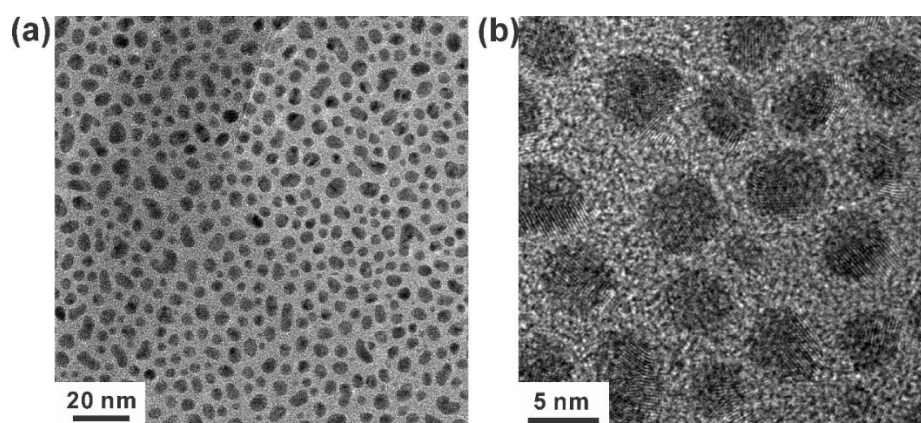


Figure S2. (a) Low-magnification TEM image and (b) HRTEM image of the Au NPs deposited by sputtering method. The lattice fringes in Au NPs can be clearly observed from HRTEM image.

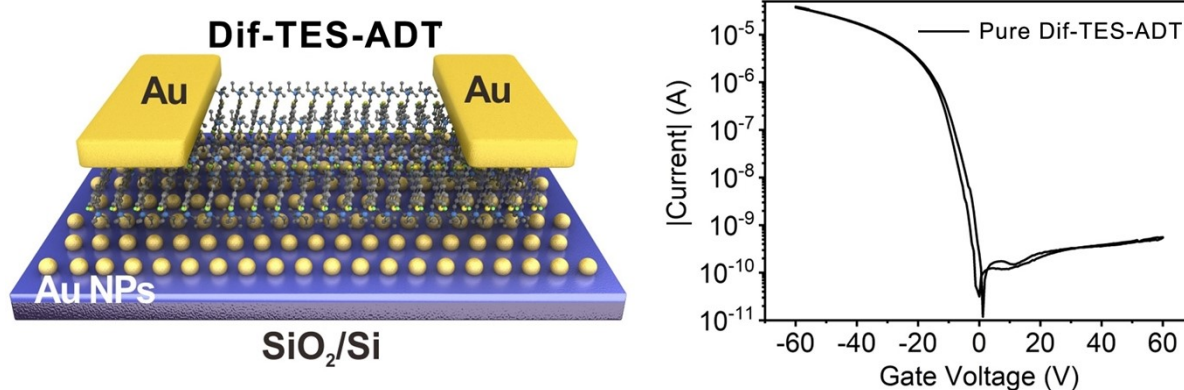


Figure S3. (a) Schematic illustration and (b) electrical transfer characteristic of a pure Dif-TES-ADT based transistor without a PS layer. A fixed V_{DS} of 1 V was applied.

The pure Dif-TES-ADT device was fabricated on the top of the flexible substrate from 3 mg mL^{-1} Dif-TES-ADT solution in toluene *via* an established blade-coating method. A piece of silicon wafer was used as the blade to drag the Dif-TES-ADT solution across the substrate at a speed of $\sim 115 \text{ } \mu\text{m s}^{-1}$. With the solvent evaporation, molecules would nucleate and crystallize, generating highly ordered Dif-TES-ADT crystals on the flexible substrate. The detailed fabrication of substrate and electrodes are the same as in the manuscript.

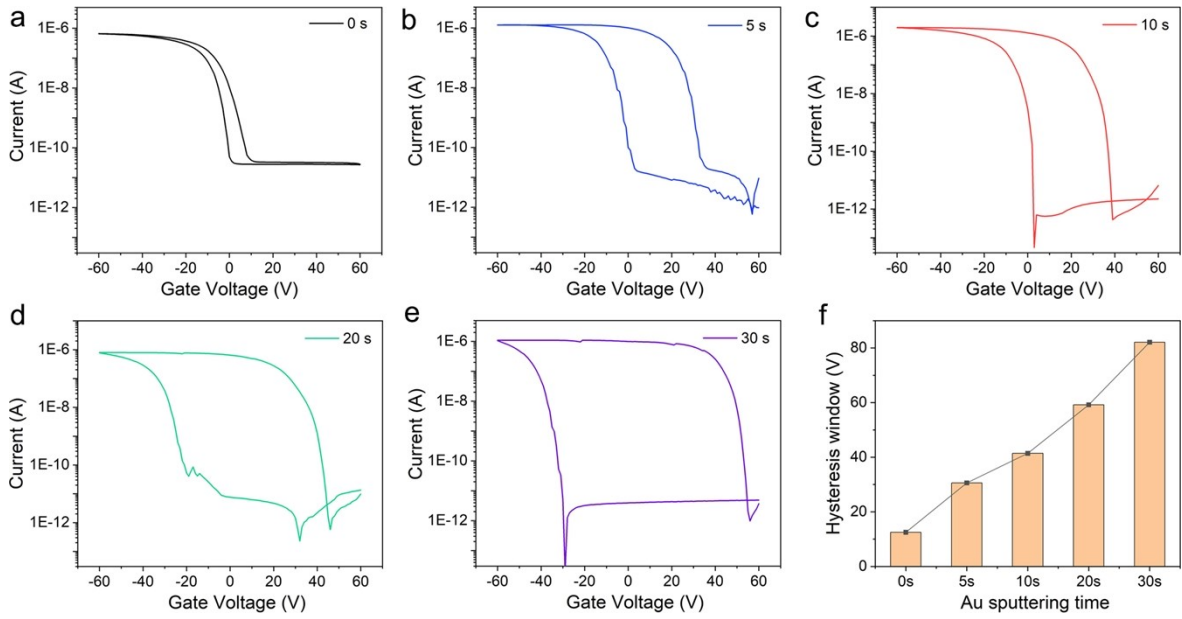


Figure S4. Electrical transfer characteristics of the FG-OFET devices with different Au NPs sputtering time of (a) 0 s, (b) 5 s, (c) 10 s, (d) 20 s, (e) 30 s and (f) the comparison of hysteresis windows in FG-OFET memory devices with different Au NPs sputtering times.

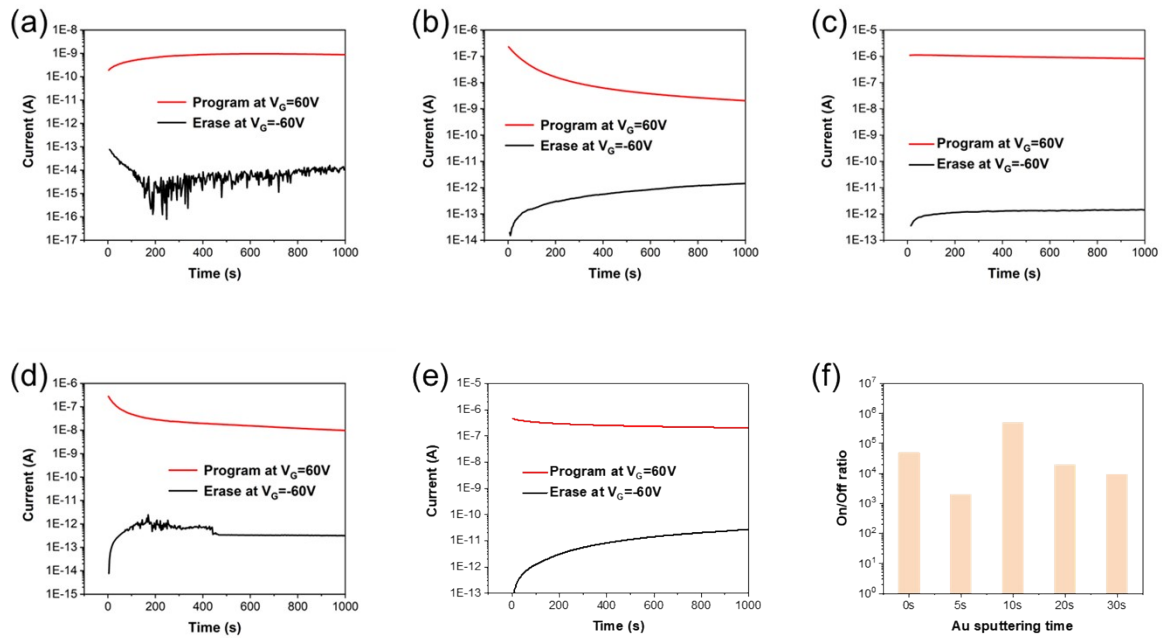


Figure S5. Retention behaviors of the FG-OFET devices with different Au NPs sputtering time of (a) 0 s, (b) 5 s, (c) 10 s, (d) 20 s, (e) 30 s and (f) the comparison of on/off ratios after 100 s.

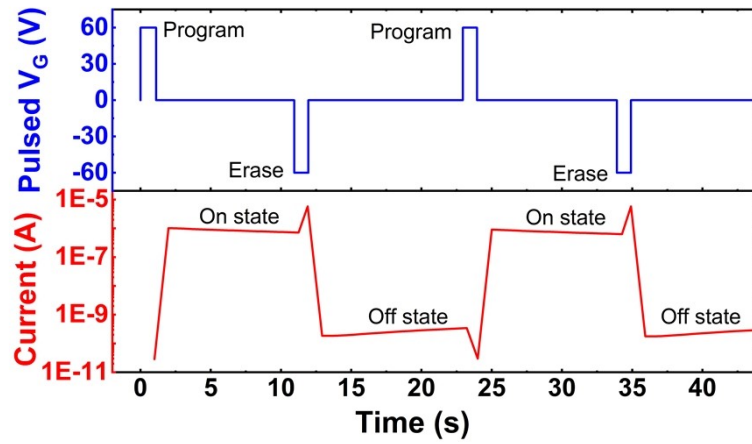


Figure S6. The detailed pattern of the pulsed gate voltage for the dynamic switching between the erased state (Off) and the programmed state (On). The duration time of the programming and erasing bias is 1 s.

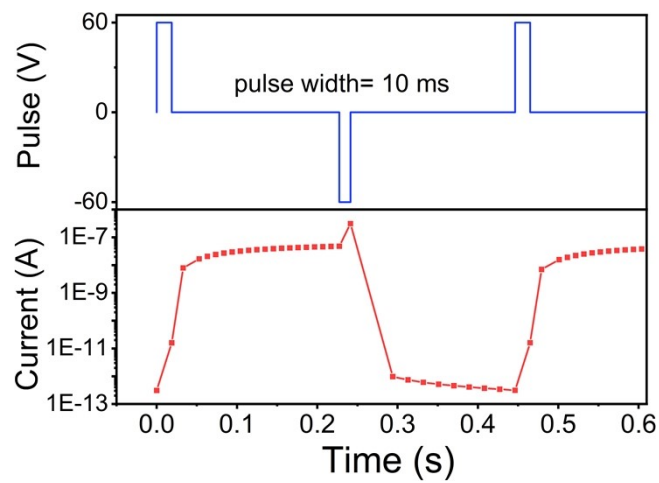


Figure S7. Transient curve of the FG-OFET memory device under the ± 60 V programming and erasing voltages with a programming/erasing pulse width of 10 ms.

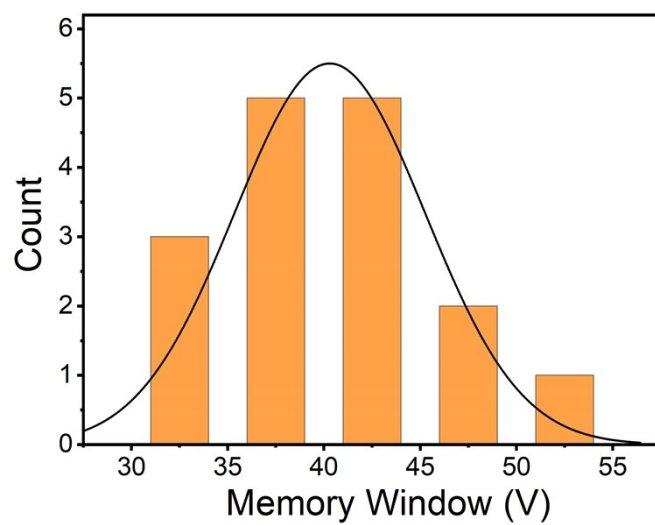


Figure S8. Memory window distribution in the 4×4 FG-OFET memory devices array on same substrate.

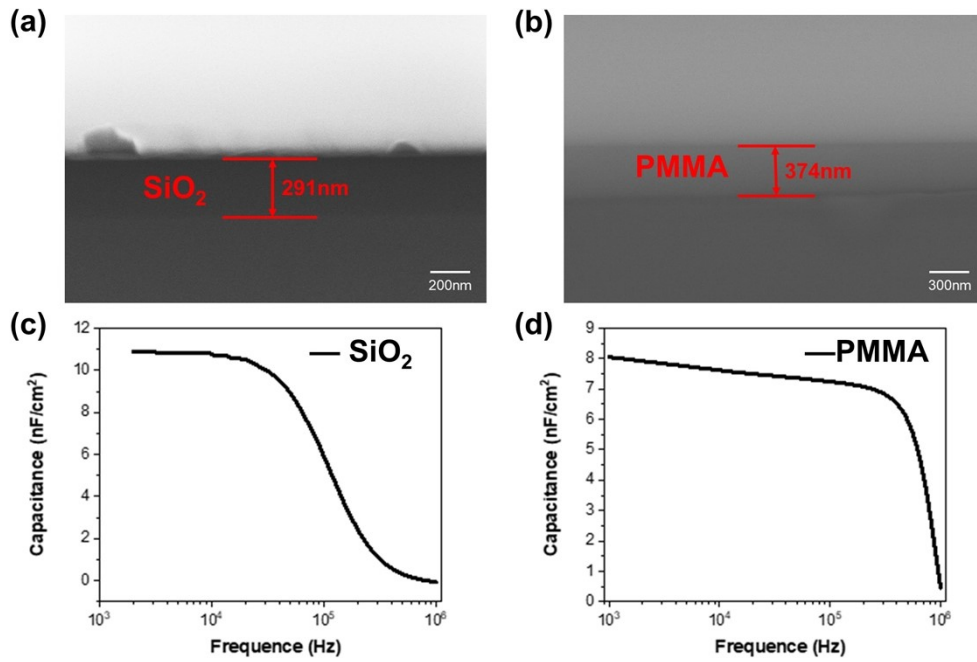


Figure S9. The cross-sectional SEM images of (a) SiO₂ and (b) PMMA dielectric layers. The capacitance per unit area-frequency characteristics of (c) SiO₂ and (d) PMMA dielectric layers.

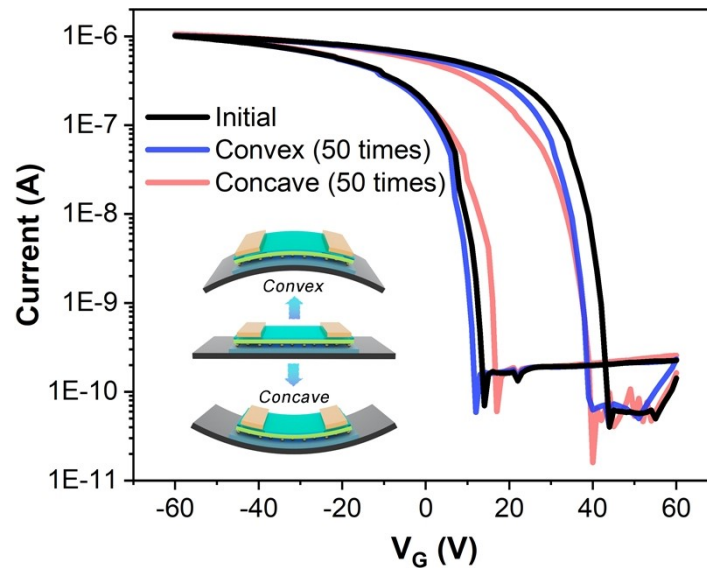


Figure S10. The endurance characteristics of the flexible FG-OFET memory device under convex and concave bending states in 50 cyclic tests. A fixed V_{DS} of 1 V was applied and the bending radius (R) was 1 cm.

Table S1. Comparison of device performances of low-dimensional semiconductor-based OFGMs.

Device structure	Hysteresis window/Operation voltage (V)	Retention (s)	ON/OFF ratio	Ref
Pentacene/SiO ₂ /Au NPs	23.6/80	N/A	~10	Ref ¹
Pentacene/AlO _x /PVDF-TrFE	20/80	2×10 ⁴	10 ³	Ref ²
Pentacene/HfO ₂	22.6/120	\	10 ³	Ref ³
Pentacene/APTES/Au NPs	10/180	10 ⁵	10 ³	Ref ⁴
P3HT/MoS ₂	23/160	\	10 ⁵	Ref ⁵
CDT-DPP-TVT/Cytop/Au NPs	25/120	10 ⁴	10 ⁴	Ref ⁶
Pentacene/Maltoheptaose	3.2/100	3.6×10 ⁴	10 ⁶	Ref ⁷
Pentacene/Polarizable polymers	33.7/140	<1000	10 ⁴	Ref ⁸
Pen/Pentafluorosulfanylated polymers	34/240	10 ⁴	10 ⁶	Ref ⁹
QQT(CN) ₄ /PVDF-TrFE	25/100	6000	<10 ³	Ref ¹⁰
DH-6T/SAM	0.1/4	~3600	2.4	Ref ¹¹
PTDPPTFT4/o-MeO-DMBI	47/200	10 ⁴	10	Ref ¹²
Dif-TES-ADT:PS/Au NPs	41.4/120	5×10 ³	10 ⁵	Our work

Reference

1. Park YS, Lee JS. Design of an efficient charge-trapping layer with a built-in tunnel barrier for reliable organic-transistor memory. *Adv. Mater.*, **27**, 706-711 (2015).
2. Xu M, Zhang X, Li S, Xu T, Xie W, Wang W. Gate-controlled multi-bit nonvolatile ferroelectric organic transistor memory on paper substrates. *J. Mater. Chem. C* **7**, 13477-13485 (2019).
3. Zhuang J, Han S-T, Zhou Y, Roy VAL. Flash memory based on solution processed hafnium dioxide charge trapping layer. *J. Mater. Chem. C* **2**, 4233-4238 (2014).
4. Kim SJ, Lee JS. Flexible organic transistor memory devices. *Nano Lett.*, **10**, 2884-2890 (2010).
5. Kang M, *et al.* Stable charge storing in two-dimensional MoS₂ nanoflake floating gates for multilevel organic flash memory. *Nanoscale* **6**, 12315-12323 (2014).
6. Jeon S, *et al.* Synthesis of cyclopentadithiophene-diketopyrrolopyrrole donor-acceptor copolymers for high-performance nonvolatile floating-gate memory transistors with long retention time. *ACS Appl. Mater. Interfaces* **12**, 2743-2752 (2020).
7. Chiu YC, Sun HS, Lee WY, Halila S, Borsali R, Chen WC. Oligosaccharide carbohydrate dielectrics toward high-performance non-volatile transistor memory devices. *Adv. Mater.*, **27**, 6257-6264 (2015).
8. Xu T, *et al.* Organic field-effect transistor nonvolatile memories with hydroxyl-rich polymer materials as functional gate dielectrics. *Adv. Electron. Mater.*, **5**, 1900569 (2019).
9. Zhang G, Lee Y-J, Gautam P, Lin C-C, Liu C-L, Chan JMW. Pentafluorosulfanylated polymers as electrets in nonvolatile organic field-effect transistor memory devices. *J. Mater. Chem. C* **7**, 7865-7871 (2019).
10. Kim RH, *et al.* Non-volatile organic memory with sub-millimetre bending radius. *Nat. Commun.* **5**, 3583 (2014).
11. Burkhardt M, *et al.* Concept of a molecular charge storage dielectric layer for organic thin-film memory transistors. *Adv. Mater.*, **22**, 2525-2528 (2010).
12. Lee WY, Wu HC, Lu C, Naab BD, Chen WC, Bao Z. n-type doped conjugated polymer for nonvolatile memory. *Adv. Mater.*, **29**, 1605166 (2017).