Highly Uniformity Photonic Synapse Arrays Based on TIPS-pentacene Nanowires/$CsPbBr_3$ heterojunctions

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Supporting Information

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Captions

Figure S1 a) the film with periodic grating patterns b)TIPS nanaowires composite synaptic array device.

Figure S2 The preparation process of $CsPbBr_3$/TIPS-pentacene composite thin films.

Figure S3 The image of $CsPbBr_3$ perovskite films on silicon wafers without doping (a) and doped with 30wt% (d) under ultraviolet light irradiation. The fluorescence microscopic morphology of $CsPbBr_3$ perovskite thin films undoped (b) - (c) and doped with 30wt% (e) - (f).

Figure S4 The AFM images of $CsPbBr_3$ perovskite films without doping (a), doped with 30wt% (b), and 50wt% (c).

Figure S5 The fluorescence microscopy morphology (a) - (b) and AFM images (c) - (d) of TIPS pentacene nanowire thin films.

Figure S6 (a) Transfer characteristic curves of $CsPbBr_3$/TIPS pentacene composite thin film transistor over multiple cycles. Extract statistical charts of the saturation mobility (b), threshold voltage (c), and current switching ratio (d) of the device from the transfer characteristic curve.

Figure S7 The photosensitivity of phototransistors based on pure TIPS films.

Figure S8 Comparison of leakage current model and experimental data of organic transistors: (a) the transfer characteristic curve and (b) the relationship between threshold voltage offset and light power.

Figure S9 The EPSC of $CsPbBr_3$/TIPS-pentacene composite synaptic device with different light pulse times (a) - (c) and different light pulse numbers (d) - (f).

Figure S10 (a) The schematic diagram of $CsPbBr_3$/TIPS-pentacene composite thin film array device. The thermal maps (b) and the statistical charts (c) of the saturation mobility. (d) Transfer characteristic curve of $CsPbBr_3$/TIPS pentacene composite thin film transistor. The thermal maps (e) and the statistical charts (f) of the threshold voltage.

Figure S11 Comparison I_{GS} and I_{DS} current curves of synaptic transistors.

Figure S12 (a) Synaptic device EPSC and its data fitting results. The thermal maps of the photocurrent time relaxation constants (b) and (c) of the synaptic array after the end of illumination.

Figure S13 The paired-pulse facilitation of synaptic transistor.

I. Compact model of organic field-effect transistors

We utilized the universal compact model of thin-film transistors (TFT) developed by Zhao et al. to simulate the transfer characteristics of a phototransistor. Under ideal conditions, the relationship between the charge density accumulated at the interface space x and the potential $V_D(x)$ along the channel direction[1, 2]:

$$Q_s(x)=n(x)et=C_{ox}\left(V_{GS}-V_{th}-V_D(x)\right) \qquad \text{Eq. (1)}$$

where $n(x)$ is the carrier concentration at x, e is the unit charge quantity, t is the thickness of the charge region in the channel, and C_{ox} is the unit capacitance of the gate dielectric layer. Obtaining the expression of current density in the channel based on Ohm's law:

$$J=ne\mu E \qquad \text{Eq. (2)}$$

Thus obtaining the relationship between device channel current and current density.

$$I_D=\frac{W}{L}\mu C_{ox}\int_0^{V_D}\left(V_{GS}-V_{th}-V_D(x)\right)dV_D(x) \qquad \text{Eq. (3)}$$

When the device operates in the linear region, $\left|V_{GS}-V_{th}\right|$? $\left|V_{DS}\right|$, the expression for leakage current is

$$\mathrm{I}_D=\frac{W}{L}\mu C_{ox}\left(V_{GS}-V_{th}-\frac{1}{2}V_D\right)V_D \qquad \text{Eq. (4)}$$

When the device operates in the saturation region, $\left|V_{DS}\right|$? $\left|V_{GS}-V_{th}\right|$, the expression for leakage current is

$$\mathrm{I}_D=\frac{1}{2}\frac{W}{L}\mu C_{ox}\left(V_{GS}-V_{th}\right)^2 \qquad \text{Eq. (5)}$$

Considering the common characteristic of the organic semiconductor trap filled conduction model or the range hopping model, which is that the mobility of thin film transistor devices depends on the gate voltage, the mobility can be expressed as[2, 3]

$$\mu_{FE} = \mu_0 \left(V_{GS} - V_{on}\right)^{\gamma} \qquad \text{Eq. (6)}$$

Taking into account the channel length modulation effect of transistors, the leakage current expression is obtained as

$$\mathrm{I}_D = \frac{W}{L} \mu_0 \left(V_{GS} - V_{on}\right)^{\gamma} C_{ox} \left(V_{GS} - V_{th} - \frac{1}{2} V_D\right) V_D \qquad \text{Eq. (7)}$$

$$\mathrm{I}_D = \frac{1}{2} \frac{W}{L} \mu_0 \left(V_{GS} - V_{on}\right)^{\gamma} C_{ox} \left(V_{GS} - V_{th}\right)^2 \left(1 + \lambda V_D\right) \qquad \text{Eq. (8)}$$

II. Theoretical estimation of charge capture effect

In optoelectronic thin film transistors based on organic/perovskite heterojunctions, there exists a type-II heterojunction relationship between the interface of organic semiconductors and perovskite materials. The perovskite near the interface absorbs light and generates photo generated electron hole pairs for rapid separation. Under the action of an internal electric field, photo generated holes quickly transfer to positions with lower energy barriers. Under steady-state conditions, the total flow rate of carriers transferred to the lowest barrier includes photogenerated carriers and carriers drifting in the dark state[4]. The hole flux in equilibrium state is expressed as

$$\Delta p_{\mathrm{light}} + \Delta p_{dark} = \Delta p_{\mathrm{all}} \qquad \text{Eq. (9)}$$

where $\Delta p_{\mathrm{light}}$ is the photogenerated carriers which related to light power P and the incident light power hv, and Δp_{dark} is the drifting carriers drifting which related to the dark state current ($\Delta p_{dark} \propto J_{pd}/q$). The accumulation of charge carriers in the active layer leads to the bending of the energy band, and the relationship between the hole concentration at the lowest point of the potential barrier and the offset of the potential barrier is[4]

$$\Delta p_{all} = C \exp\left(\frac{\Delta \varphi_b}{n\left(kT/q\right)} \right) \qquad \text{Eq. (10)}$$

where C is a proportional parameter and n is a constant that describes the saturation effect under high current. Then we obtain the expression for the potential barrier offset caused by photogenerated carriers as

$$\Delta\varphi_b = \frac{n_1 kT}{q}\ln\left(1+\frac{q\cdot n_2\cdot \Delta p_{light}}{J_{pd}}\right) \qquad \text{Eq. (11)}$$

where n_1 and n_2 are proportional parameter.The threshold voltage of thin film transistors describes the gate voltage at which the channel begins to form conductive current. When additional photo generated carriers accumulate in the channel, resulting in the bending of the interface energy band, the channel is more likely to form an effective conductive channel. The offset of the threshold voltage is positively correlated with the change in the interface potential barrier[5] ($\Delta V_{th} = n\Delta\varphi$). The relationship between the threshold voltage offset a based on heterojunction thin film transistors and the incident light power b is[5, 6]

$$\Delta V_{th} = \frac{n_1 kT}{q}\ln\left(1+\frac{q\cdot \eta_1\cdot P}{J_{pd}\cdot hv}\right) \qquad \text{Eq. (12)}$$

where η_1 is a coefficient related to quantum yield.

Under illumination conditions, semiconductors absorb light to generate photogenerated electron-hole pairs, and the production rate of photo generated excitons is $G = \alpha\beta P/hv$, where α is the absorption coefficient of the semiconductor, β is the quantum yield. The recombination rate of charge carriers is determined by the concentration and lifetime of non equilibrium charge carriers ($R = \Delta p/\tau$). Considering the dominance of photogenerated carriers under lighting conditions, there is a relationship between the net increase in photogenerated charge carriers generated after semiconductor absorption:

$$\frac{d(\Delta p)}{dt} = G - R = \alpha\beta\frac{P}{hv} - \frac{\Delta p}{\tau} \qquad \text{Eq. (13)}$$

Using the starting condition ($t = 0$, $\Delta p = 0$), we obtain the equation to obtain the net increase in photogenerated carrier concentration as

$$\Delta p = \alpha\beta \frac{P}{hv} \tau \left(1 - \exp\left(-t/\tau\right)\right) \qquad \text{Eq. (14)}$$

When the light stops, we similarly obtain a net photogenerated carrier concentration of

$$\Delta p = \alpha\beta \frac{P}{hv} \tau \exp\left(-t/\tau\right) \qquad \text{Eq. (15)}$$

When an organic phototransistor operates in a transient state, the current model is

$$I = J \cdot W = q \cdot \Delta p \cdot \mu \cdot W \cdot \frac{dV_{ds}(x)}{dx} \qquad \text{Eq. (16)}$$

Considering the relationship between photogenerated carrier concentration and time during the illumination process, the expression for the relationship between transient current and time under light stimulation is

$$I_{light} = q \frac{W}{L} \frac{P\alpha\beta}{hv} V_{ds} \tau \left(1 - \exp\left(-t/\tau\right)\right) \qquad \text{Eq. (17)}$$

The transient current during the photocurrent relaxation process after the cancellation of illumination is

$$I_{after,light} = q \frac{W}{L} \frac{P\alpha\beta}{hv} V_{ds} \tau \exp\left(-t/\tau\right) \qquad \text{Eq. (18)}$$

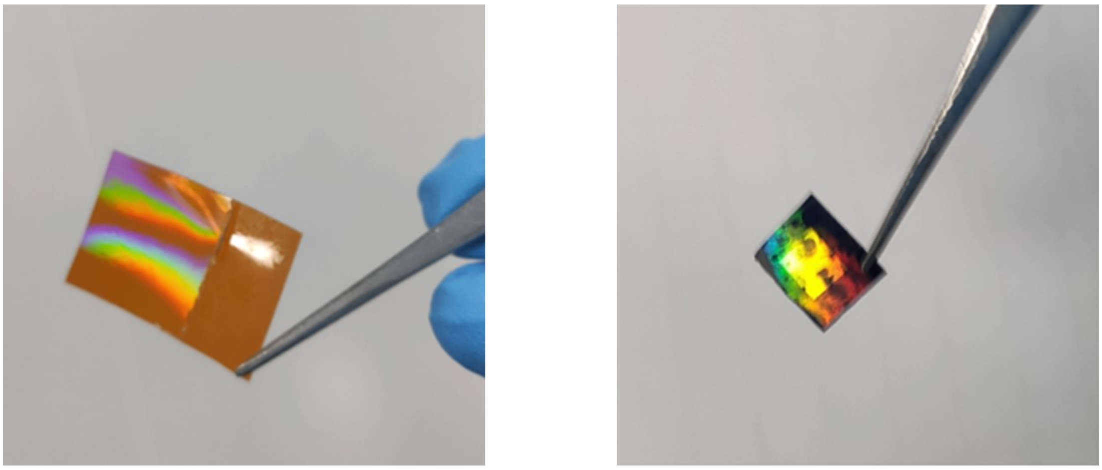
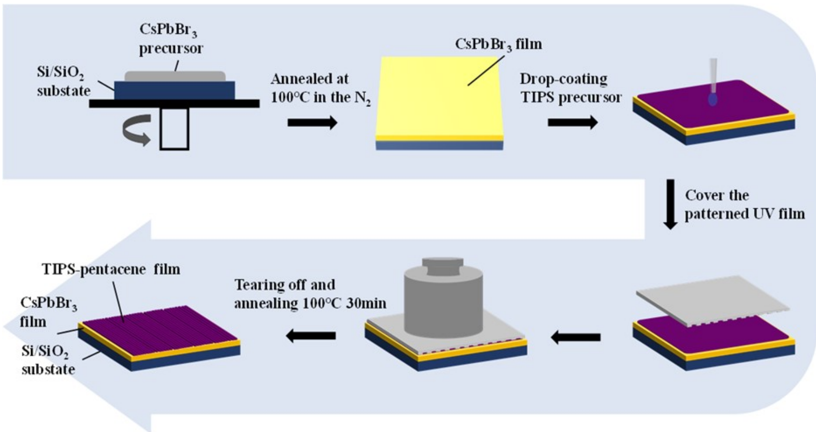


Figure S1



CsPbBr3 precursor
Si/SiO2 substate
Annealed at 100°C in the N2
CsPbBr3 film
Drop-coating TIPS precursor
Cover the patterned UV film
Tearing off and annealing 100°C 30min
TIPS-pentacene film
CsPbBr3 film
Si/SiO2 substate

Figure S2

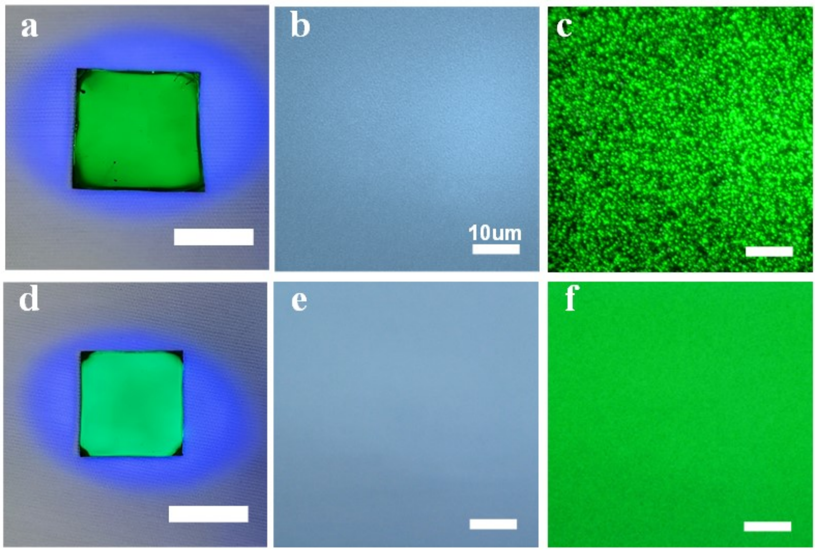


Figure S3

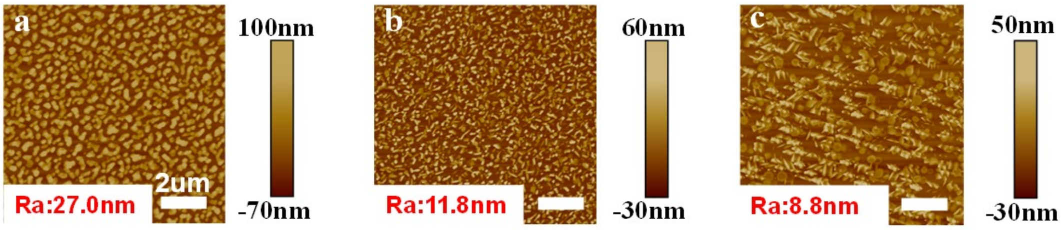

a
100nm
2um
Ra:27.0nm
-70nm
b
60nm
Ra:11.8nm
-30nm
c
50nm
Ra:8.8nm
-30nm

Figure S4

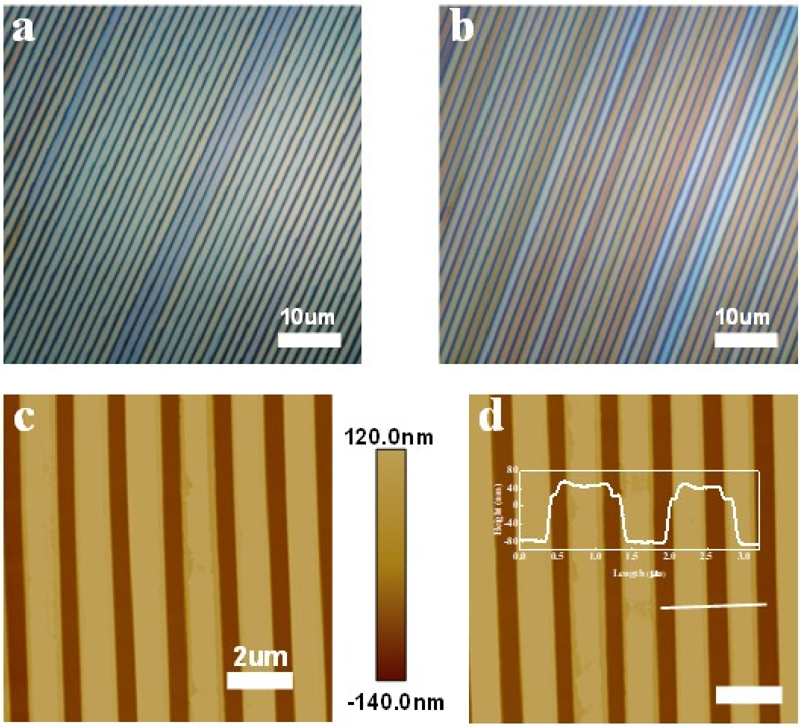


Figure S5

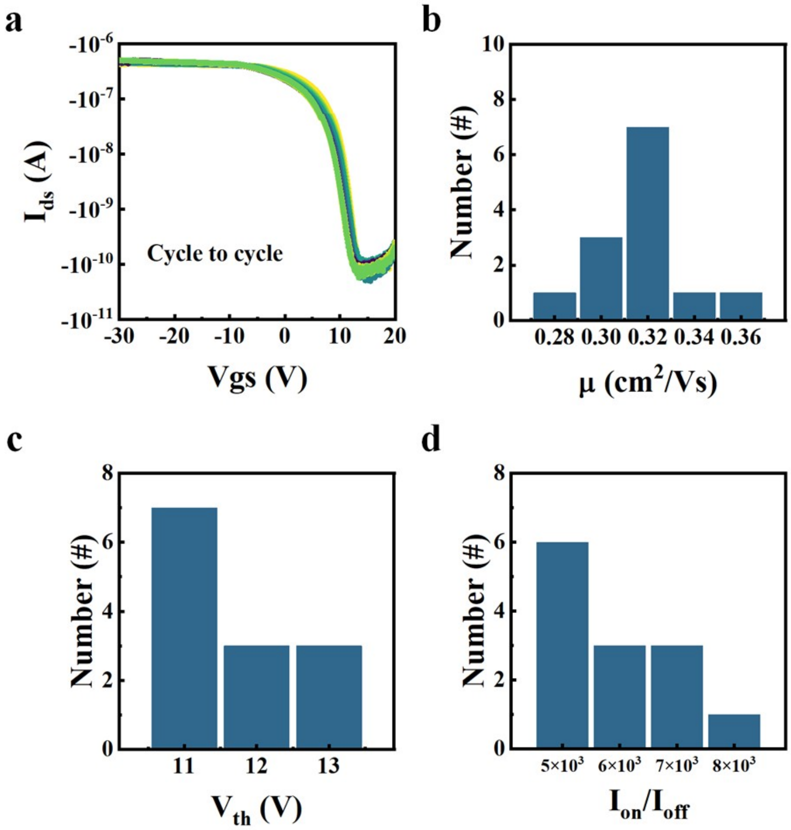
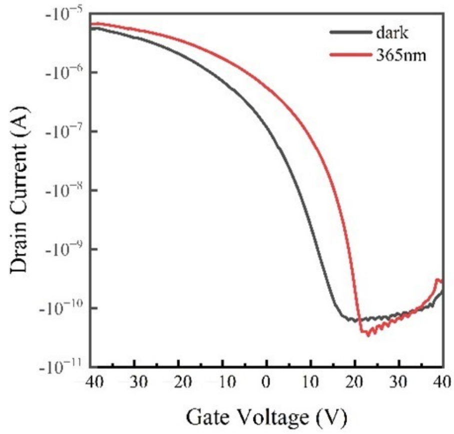

a
-10^{-6}
-10^{-7}
-10^{-8}
-10^{-9}
-10^{-10}
10^{-11}
I_{ds} (A)
Cycle to cycle
-30 -20 -10 0 10 20
Vgs (V)
b
Number (#)
0 2 4 6 8 10
0.28 0.30 0.32 0.34 0.36
μ (cm^2/Vs)
c
Number (#)
0 2 4 6 8
11 12 13
V_{th} (V)
d
Number (#)
0 2 4 6 8
5×10^3 6×10^3 7×10^3 8×10^3
I_{on}/I_{off}

Figure S6



dark
365nm
Drain Current (A)
Gate Voltage (V)
-10^−5
-10^−6
-10^−7
-10^−8
-10^−9
-10^−10
-10^−11
40
30
20
10
0
10
20
30
40

Figure S7

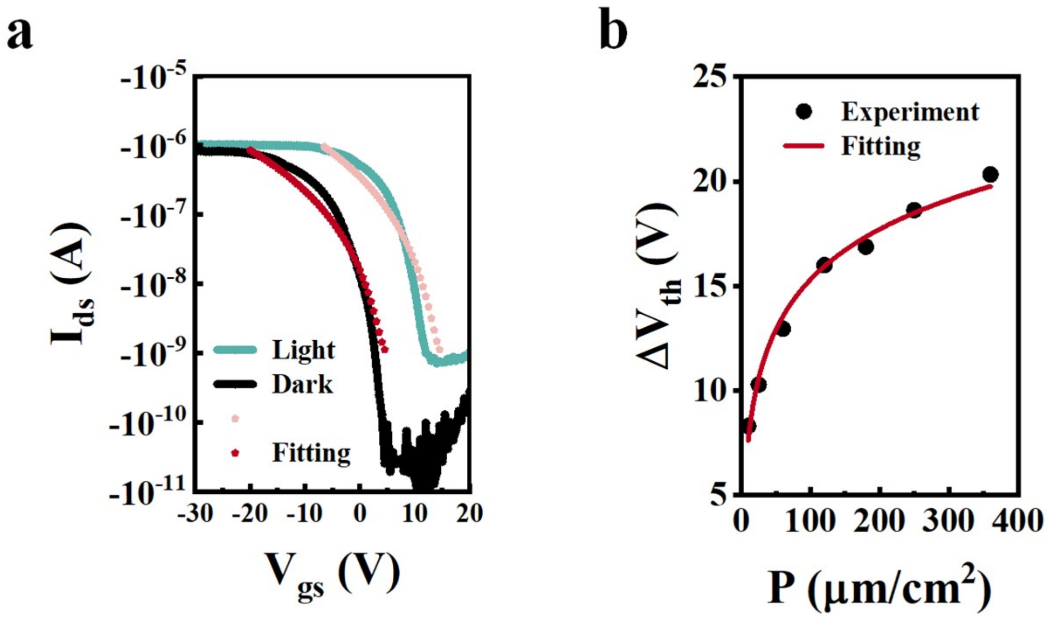

a
b
-10^-5
-10^-6
-10^-7
-10^-8
-10^-9
-10^-10
-10^-11
I_ds (A)
Light
Dark
Fitting
-30
-20
-10
0
10
20
V_gs (V)
25
20
15
10
5
ΔV_th (V)
Experiment
Fitting
0
100
200
300
400
P (μm/cm^2)

Figure S8

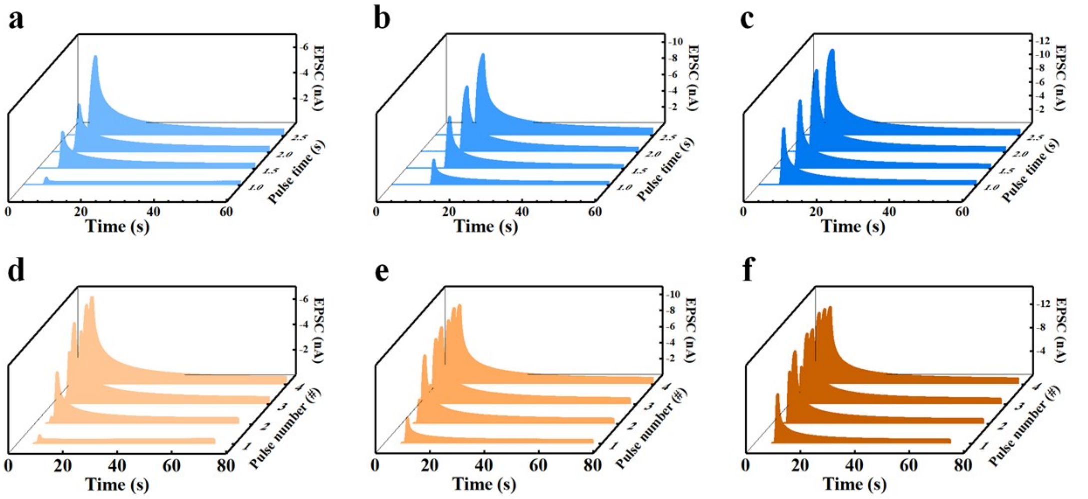
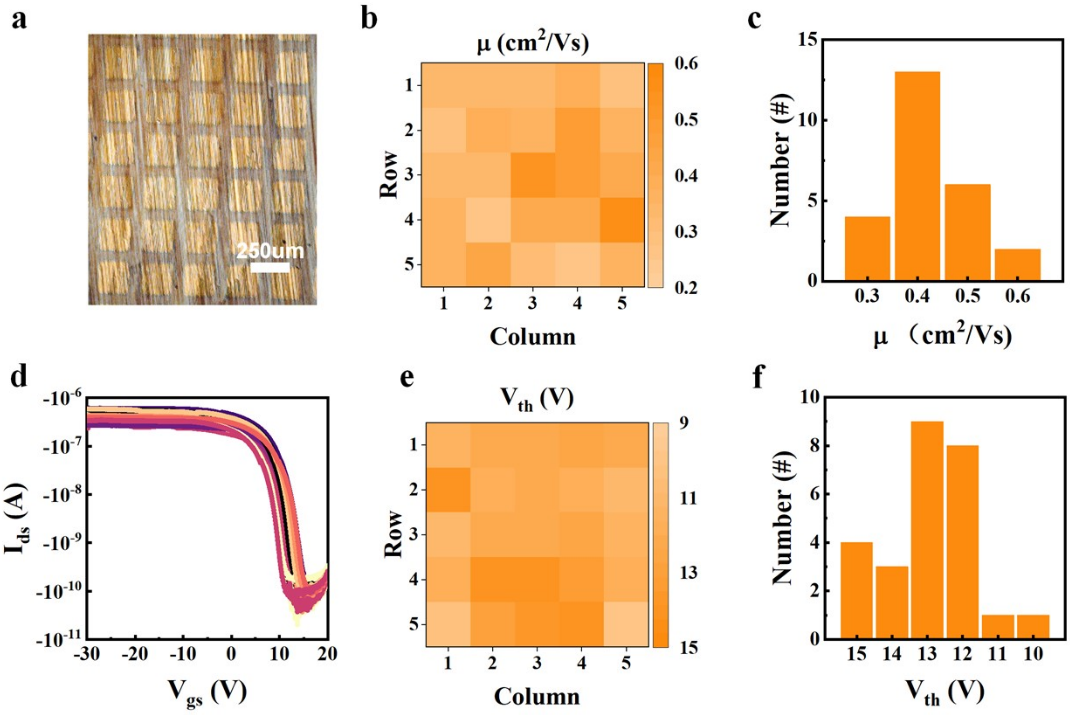
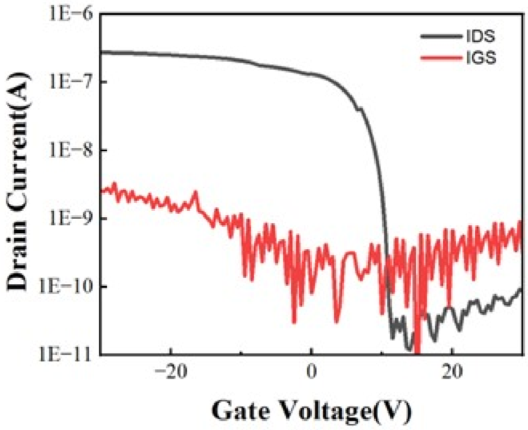


Figure S9



a
250um
b
μ (cm²/Vs)
Row
Column
c
Number (#)
μ (cm²/Vs)
d
I_{ds} (A)
V_{gs} (V)
e
V_{th} (V)
Row
Column
f
Number (#)
V_{th} (V)

Figure S10



IDS
IGS
1E−6
1E−7
1E−8
1E−9
1E−10
1E−11
Drain Current(A)
−20
0
20
Gate Voltage(V)

Figure S11

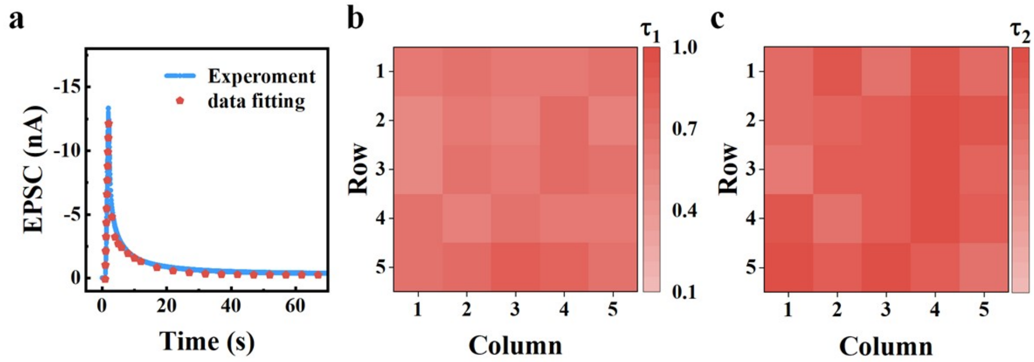

a
EPSC (nA)
Experoment
data fitting
Time (s)
b
τ_1
Row
Column
c
τ_2
Row
Column

Figure S12

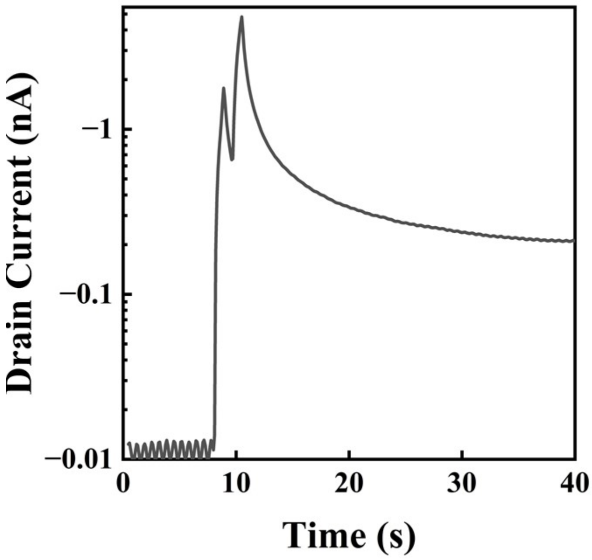


Figure S13

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