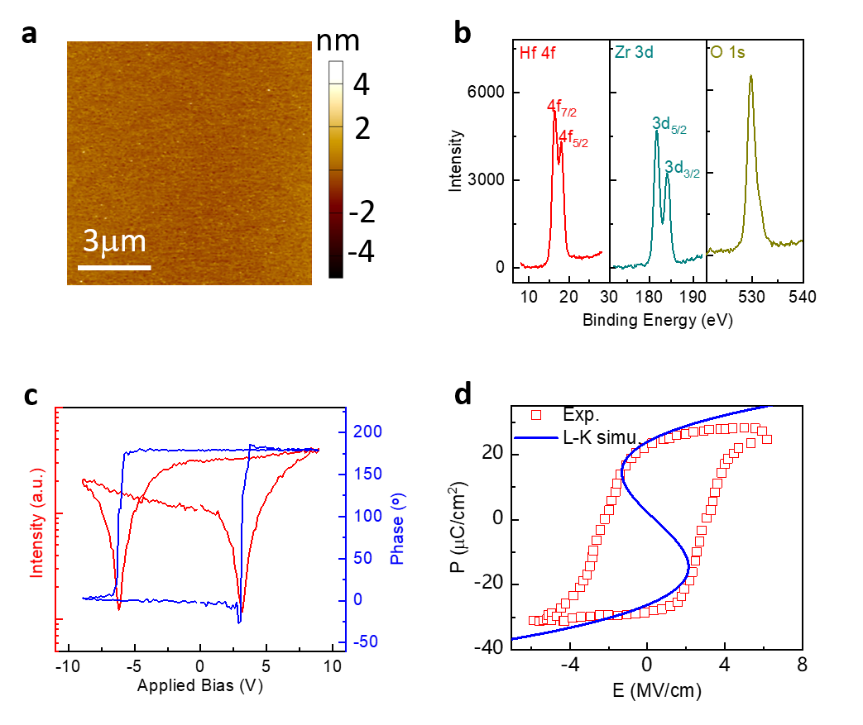
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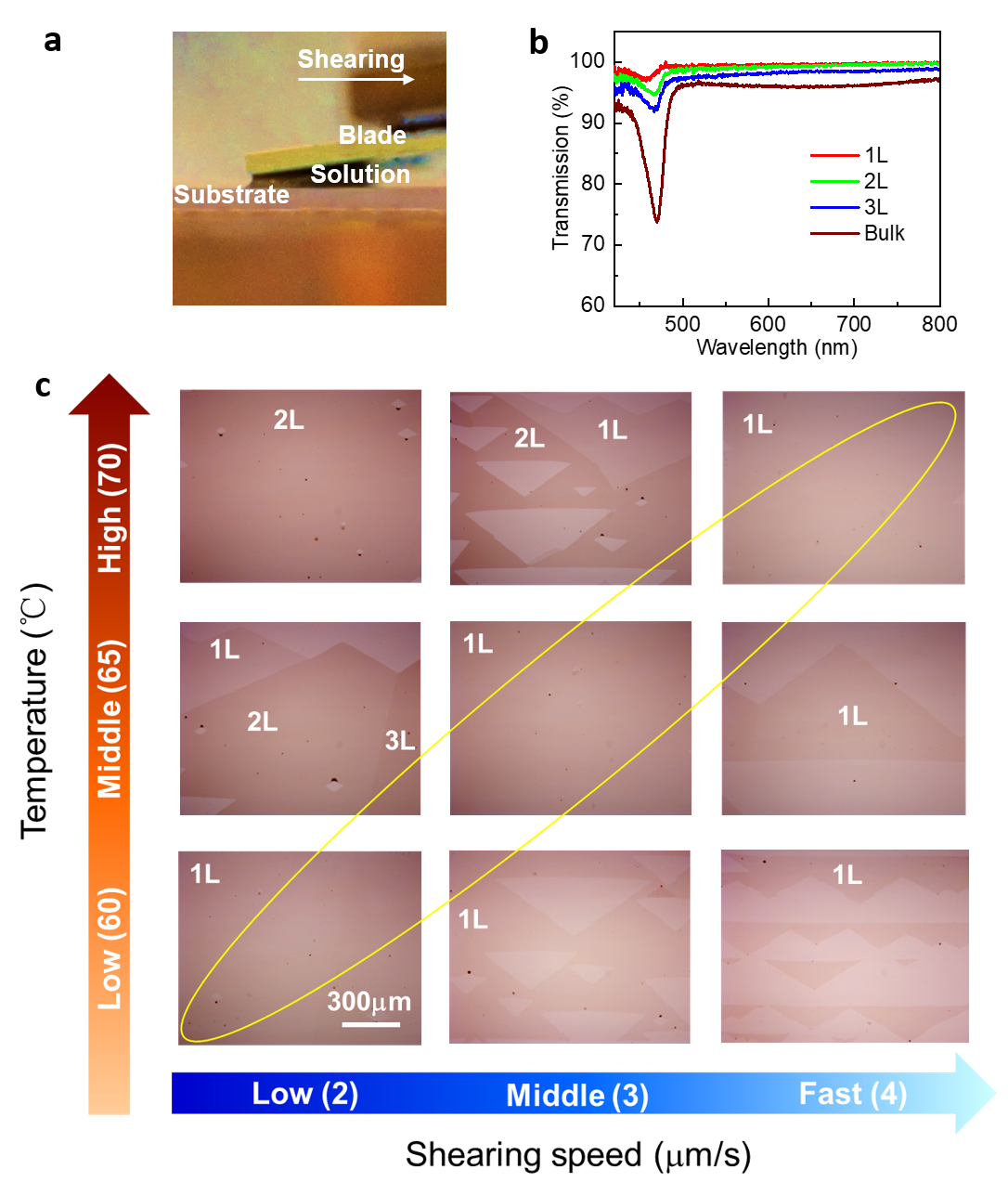
**Sub-thermionic, ultra-high-gain organic transistors and circuits**

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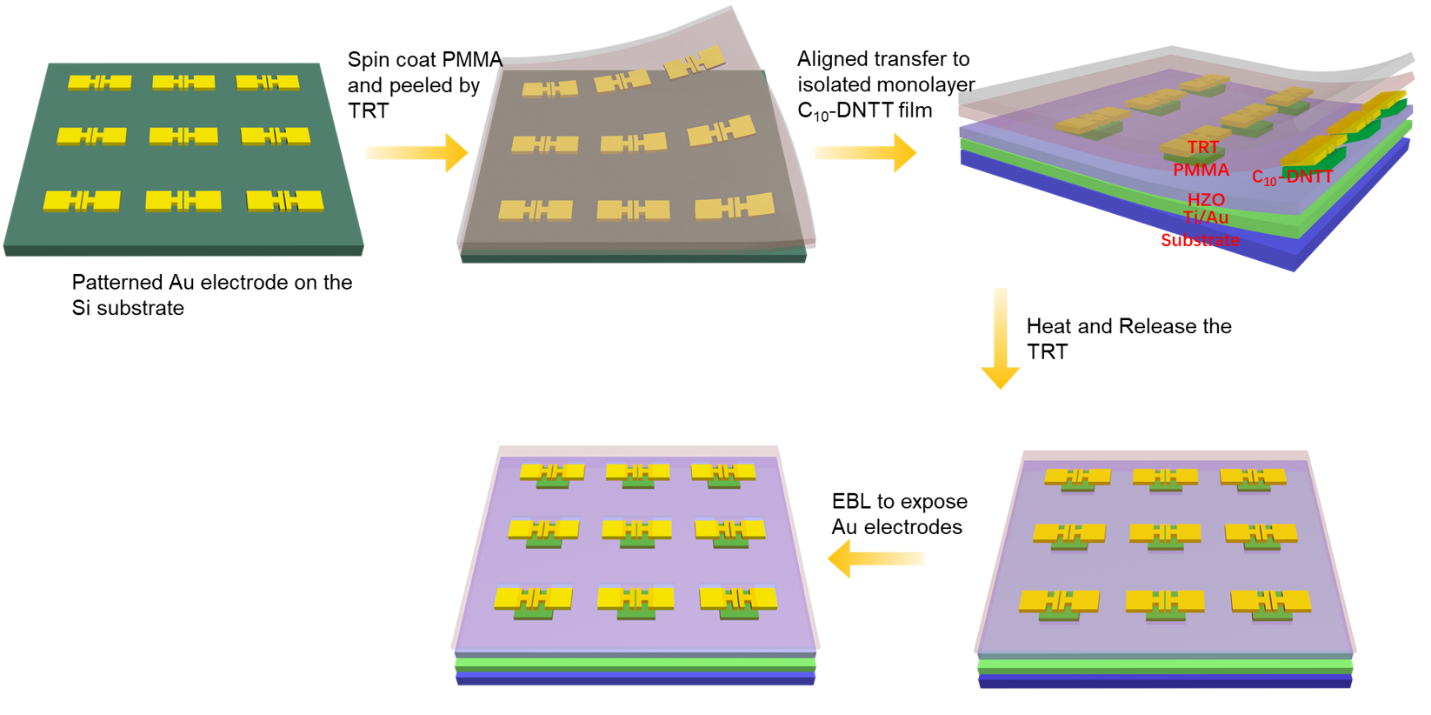
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Supplementary Figure 1| Characterization of HZO dielectric. a, AFM image and b, XPS of the HZO film on Si substrate. The composition of Hf:Zr = 1:0.95. c, Single-point PFM characterization showing ferroelectric behavior of HZO. d, The experiment Polarization-electric field loop of Si/ 22 nm HZO/Au stack showing hysteretic behavior (red square). The blue line is the fitting curve using the L-K equation to extract the Landau coefficients.

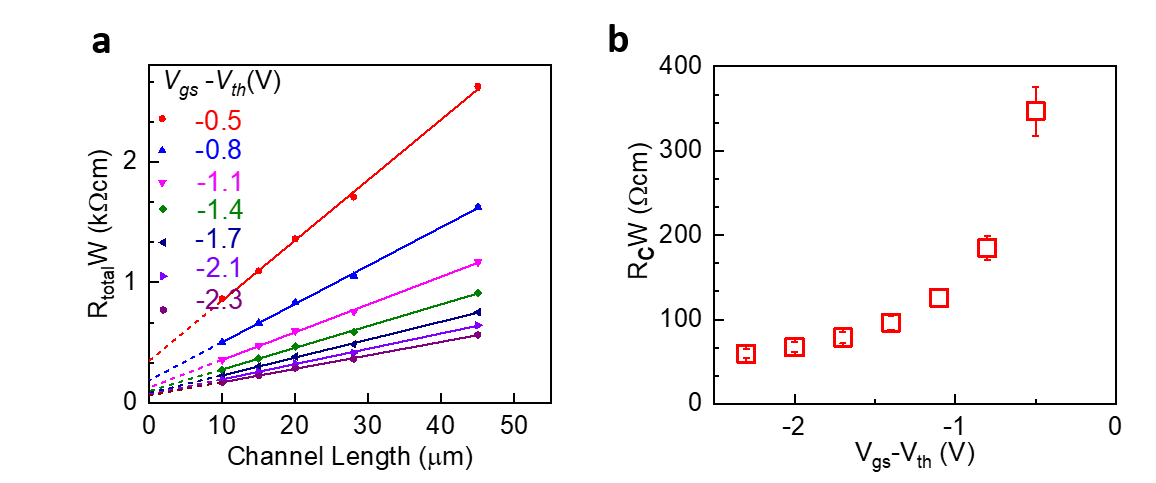


Supplementary Figure 2| Solution shearing of C10-DNTT films. a, Photograph of the shearing process. b, Transmission spectra of C10-DNTT films at different number of layers. c, The film morphology depends on the substrate temperature and shearing speed. By optimizing the growth condition, uniform and large area monolayer could be realized.

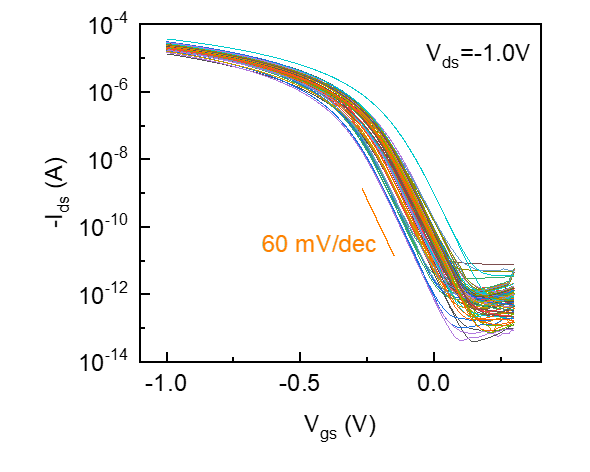


Supplementary Figure 3| Schematic illustration of vdW fabrication process for sub-thermionic OTFTs array.

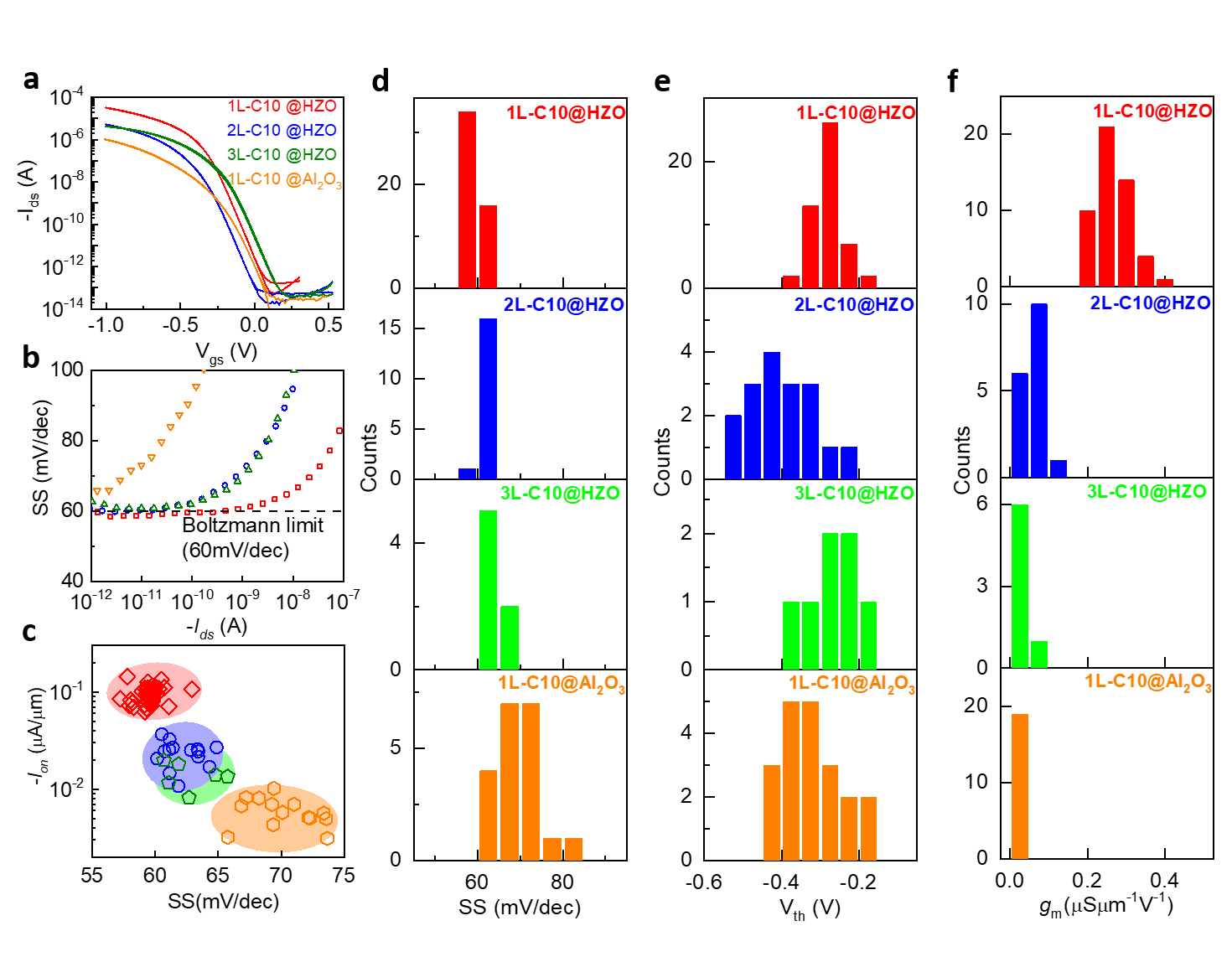
1. Fabricate the patterned Au electrode on the Si substrate using EBL and EBE;
2. Spin coat PMMA and then peel the electrode and PMMA using thermal released tape (TRT);
3. Aligned transfer to patterned monolayer C10-DNTT film;
4. Heat at 90 ℃ to release the TRT;
5. Perform EBL to pattern the PMMA to expose Au electrodes;



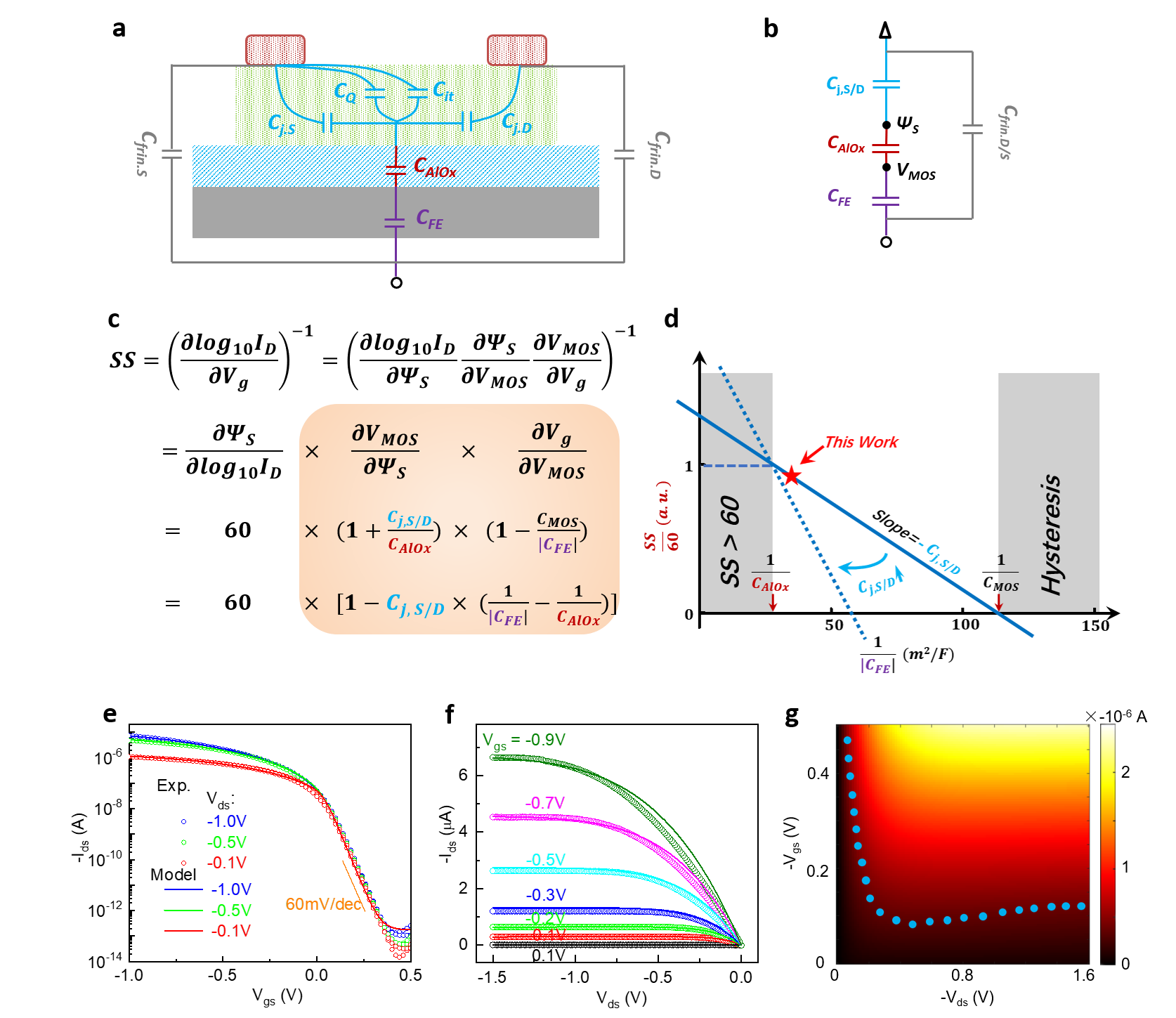
Supplementary Figure 4| Contact resistance measurement. a, Linear fits to the total width-normalized resistance (*R*total*W*) at different gate overdrive voltages. b, Width-normalized contact resistance (*R*C*W*) plotted as a function of the gate-overdrive voltage.



Supplementary Figure 5| Transfer characteristics of 50 sub-thermionic monolayer OTFTs.



Supplementary Figure 6| Thickness and substrate dependence of the OTFTs. a, *I*ds-*V*gs characteristics of typical monolayer, bi-layer and tri-layer C10-DNTT OTFTs on 22 nm HZO/2 nm Al2O3 as well as monolayer C10-DNTT OTFT on 24 nm Al2O3 under *V*ds = -1 V. b, *SS* versus *I*ds characteristics of the devices in a. c, Statistical analysis of *I*on vs *SS* for monolayer (red), bi-layer (blue) and tri-layer (green) C10-DNTT OTFTs on 22 nm HZO/ 2nm Al2O3 as well as monolayer C10-DNTT OTFTs on 24 nm Al2O3 (orange). *I*on is defined as the current at |*V*ds| = |*V*gs(on)-*V*gs(off)| = 1 V. Statistical distribution of *SS* (d), *V*th (e) and normalized (f) for different thickness and substrates.



Supplementary Figure 7| Device model of sub-thermionic monolayer OTFT. a, Capacitors network and b, simplified small-signal model of sub-thermionic OTFTs. c, Formulating *SS* by decoupling the electrostatics and transport contributions. d, Graphical display of the dependence of *SS* on capacitance. The region of is the design space for capacitance matching, where SS < 60 mV/dec without hysteresis. In the left shade region (), *SS* > 60 mv/dec. In the right shade region (), *SS* < 0, hysteresis appears. The dashed line illustrates the ultra-small window for capacitance matching, when the *C*j,S/D increases. e,f, Experimental transfer and output characteristics (symbols) and the model fitting result (solid line). The model successfully reproduces the experimental data. g, Calculated output curves at different *V*gs and the blue points are *I*ds = -0.5 μA, showing negative to positive differential resistance crossover, which is responsible for the very large *r*0.

The ferroelectric (FE) properties of HZO can be described as double-well energy landscape by the Landau Khalatnikov (L-K) equation. According to the L-K equation, in the interval between coercive fields, the value of differential capacitance *C*FE is the negative, which is also metastable state regime. Through proper capacitance matching, we can stabilize the metastable negative capacitance (NC) range, and the NC effect can be used to realize sub-thermionic transport. The Landau coefficients can be extracted by fitting the experimental P-E curve of HZO ferroelectric capacitor to calculate the ferroelectric capacitance *C*FE. Figure. S1D shows the best fitting results through L-K equation with and , and the corresponding .

We extend the model in Ref. 1 for quantitative analysis and modify a physical based NC-FET analytical model2 for our 2D organic monolayer structure, incorporating the multi-domain L-K theory with the domain interaction term, polarization relaxation and semi-classical Boltzmann transport theory. To predict the characteristics of the sub-thermionic OTFT and understand more details of the mechanisms, we depict all relevant capacitors in our back-gate sub-thermionic OTFT in Supplementary Fig. 7a. *C*FE and *C*AlOx are the capacitance of HZO layer and Al2O3 layer, respectively. *C*Q, *C*it, and *C*j,S/D represent quantum capacitance (negligible in subthreshold regime of OTFT), interface trap induced capacitance (negligible due to the small value), and capacitance of Schottky junction under source/drain electrodes, respectively. Therefore, (where is dielectric constant of organic layer, which is about 33, and is thickness of Schottky junction4) is the dominant capacitance in the channel region. Due to the low doping concentration of the ultra-thin channel, is close to the monolayer molecular thickness of 4 nm, and the corresponding . In addition, *C*AlOx is measured to be Supplementary Fig. 7b plots the simplified small-signal model of sub-thermionic OTFTs. It is worth noting that source/drain-to-gate fringing capacitance *C*frin is connected in parallel outside the capacitor network that modifies channel charges, and does not contribute to capacitor matching. Based on the analytical model in Supplementary Fig. 7c, we can plot the relationship of *SS* versus 1/|*C*FE| in Supplementary Fig. 7d. To satisfy non-hysteretic conditions, 1/|*C*FE| need to be smaller than 1/*C*MOS (1/*C*MOS = 1/*C*j,S/D +1/*C*AlOx), while to maintain *SS* < 60mV/dec, 1/|*C*FE| need to be greater than 1/*C*AlOx.|*C*FE | in the optimal NC-FET should fall between *C*AlOx and *C*MOS. Therefore, as shown in Supplementary Fig. 7d, reduced *C*j,S/D will enlarge the |*C*FE| design space significantly. Meanwhile, small *C*j,S/D appears as smaller slope in Supplementary Fig. 7d, which will make it more difficult to reduce *SS* because extremely thick ferroelectric will be needed.

It is essential to extract the key parameters using capacitor network above in real device after setting up the whole physical model of sub-thermionic OTFT, to guarantee the reasonable values. The surface potential is the clue of the whole model, obtained by solving the voltage balance condition equation in the vertical direction of sub-thermionic OTFT via the special method 2. The voltage across the FE layer, , is obtained by L-K theory, as Eq. (1).

() (1)

The relationship for and in the Landau theory is a cubic function. and can be obtained via the two poles. The third and fourth items in the right part of Eq. (1) represents depolarization field5 and the last one is multi-domain interaction item6, both of which modulated severely the negative differential conductance (NDC) effect and well considered as the most possible reason of NC effect. and are the Landau coefficients relating to remnant polarization and coercive field. is the thickness of FE layer, and are the depolarization and domain interaction coefficients. The gate charge , which is obtained by the Poisson Equation and modified as areal density for monolayer C10-DNTT, is equivalent to Polarization when the effect of multi-domain, depolarization has already incorporated as the extra items.

Based on the continuity equation, *I*-*V* characteristics can be established. The Current at any position in the channel comprising of drift and diffusion components is given by Eq (2)-Eq (3), with mobility model considering the scattering mechanisms and temperature dependence7 in Eq (4).

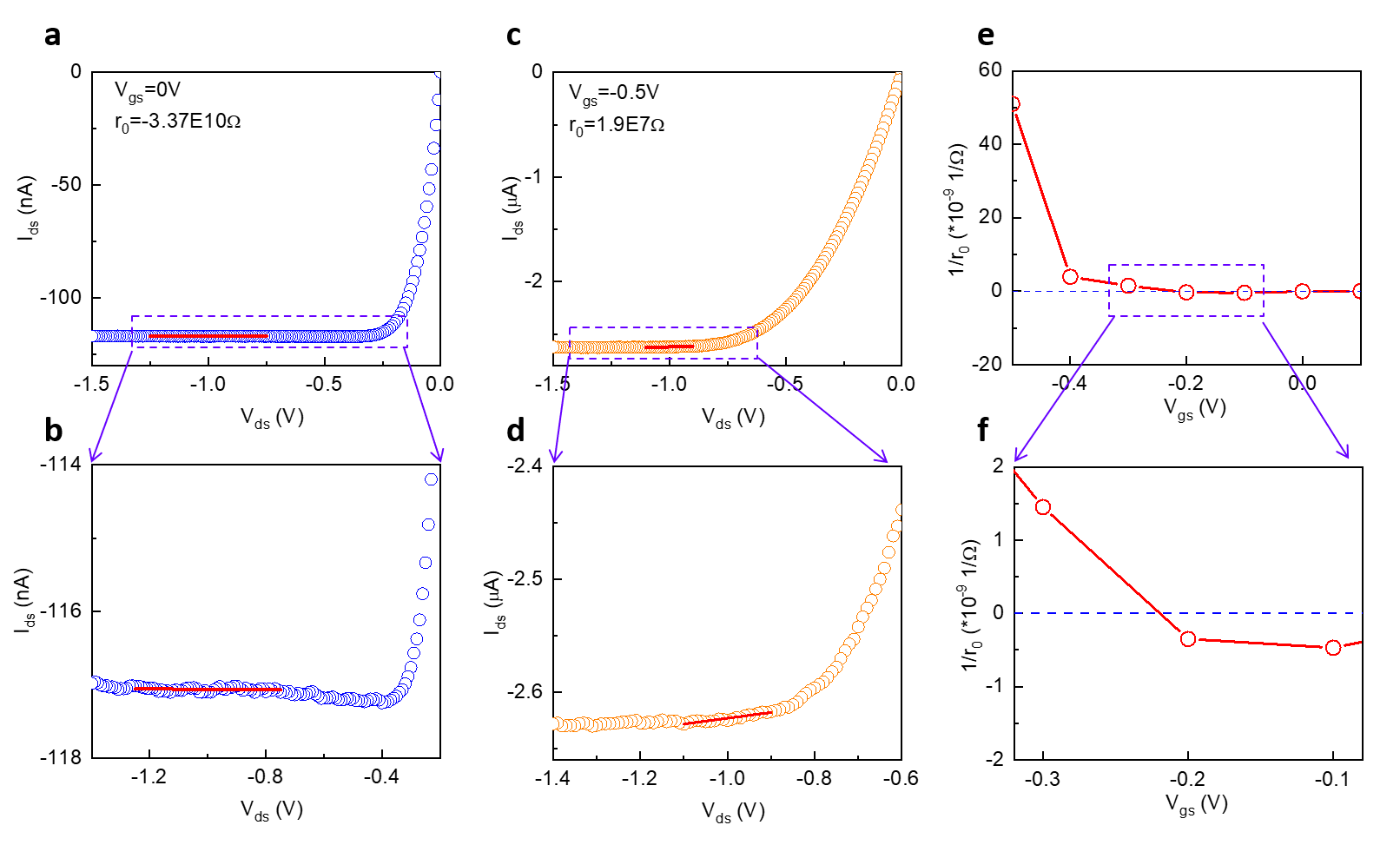
(2)

(3)

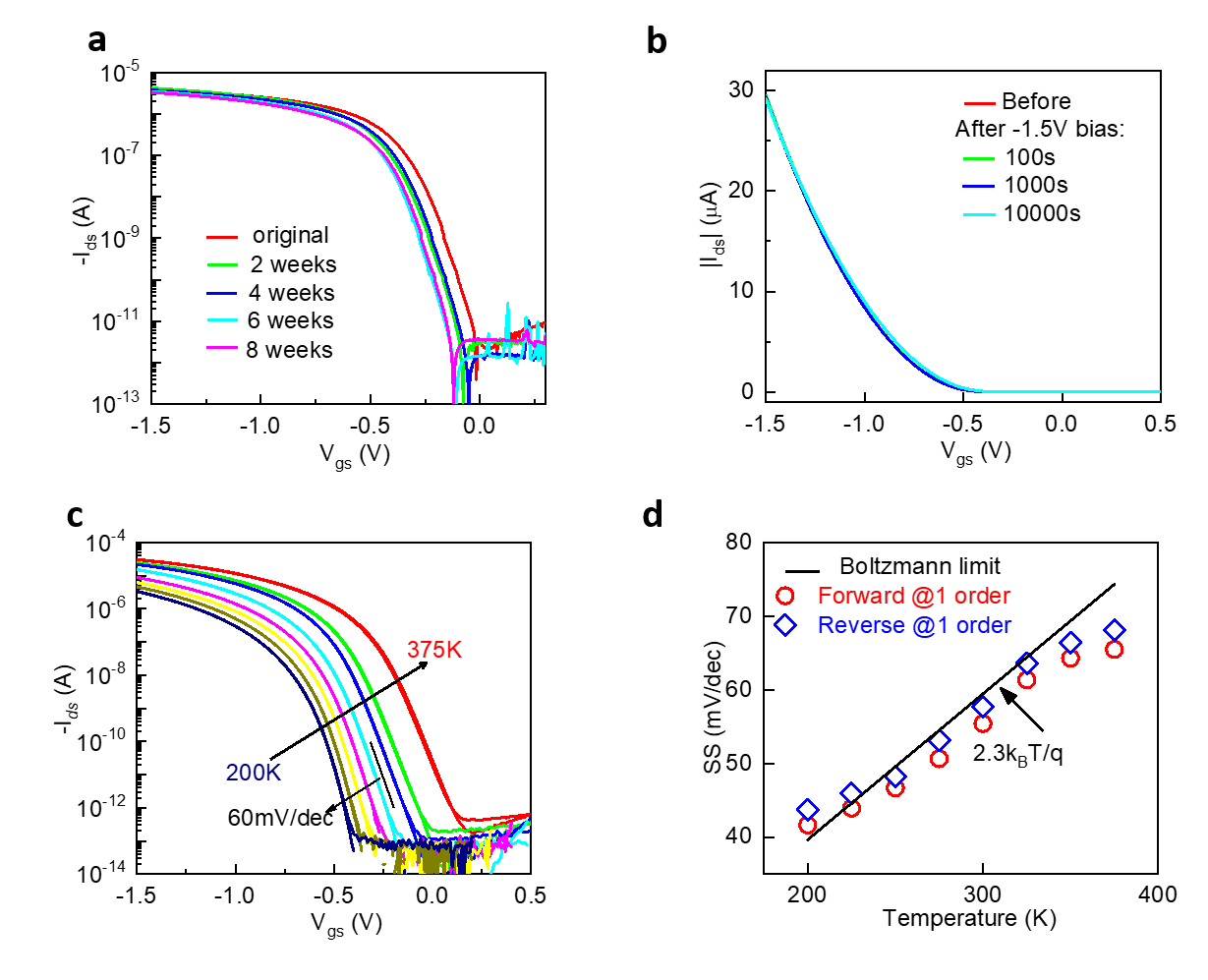
(4)

The parameter including some intrinsic parameters like permittivity, unit charge, body factor. , and are the width, length of channel and capacitances of Al2O3 layer, respectively. is the thermal voltage and is the effective average transverse electric field in channel, is the originate mobility, while , and are the Semi-fitting parameters representing the strengthen of scattering mechanism.

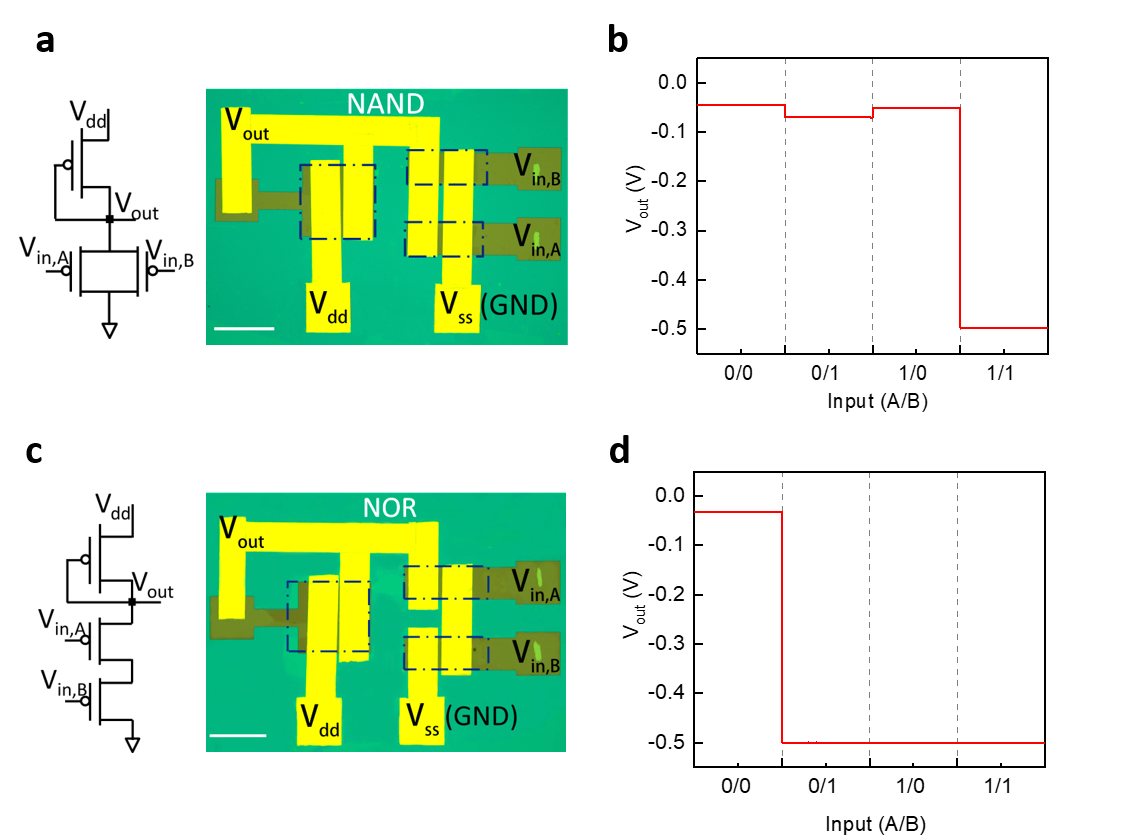
The current equation can be simplified in linear region, and the originate mobility and other parameters like can be extracted from the experimental data. An accurate match between prediction results of the model and experimental data for transfer (in linear and log coordinates) and outputcharacteristics are given in Supplementary Fig. 7e,f, via fine tuning the value of parameters in a reasonable tiny range. Supplementary Fig. 7g plots a mapping diagram of simulated *I*ds versus *V*ds and *V*gs, where the blue points are *I*ds = -0.5 μA, and the NDC can be clearly captured.



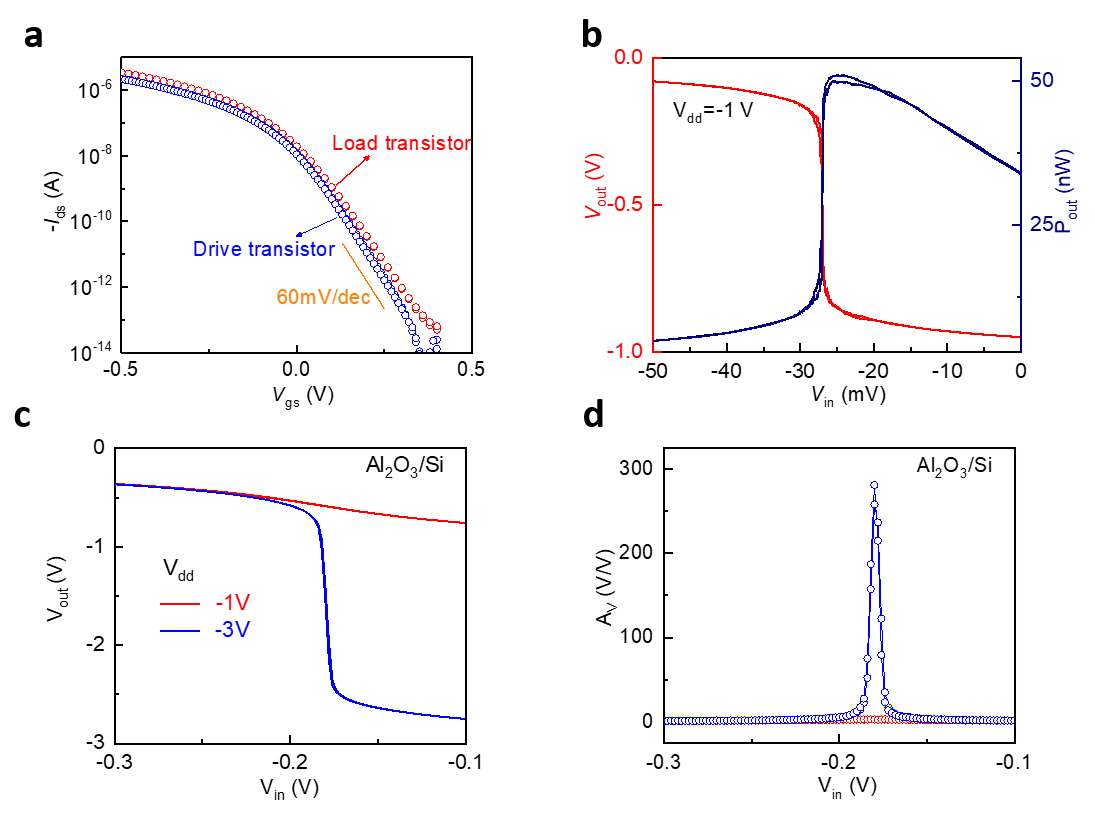
Supplementary Figure 8| Output resistance of the sub-thermionic OTFTs. a-d, Output characteristics at *V*gs = 0 and -0.5 V of a sub-thermionic OTFT. When *V*gs = 0 V, the negative differential resistance (NDR) phenomenon appears and the output resistance is negative; when *V*gs = -0.5 V, the NDR disappears and the output resistance is positive. The red curve is the linear fitting to extract the output resistance. e, Output resistance as a function of *V*gs. f, Zoom-in of the curve in e, showing a transition of output resistance from negative to positive.



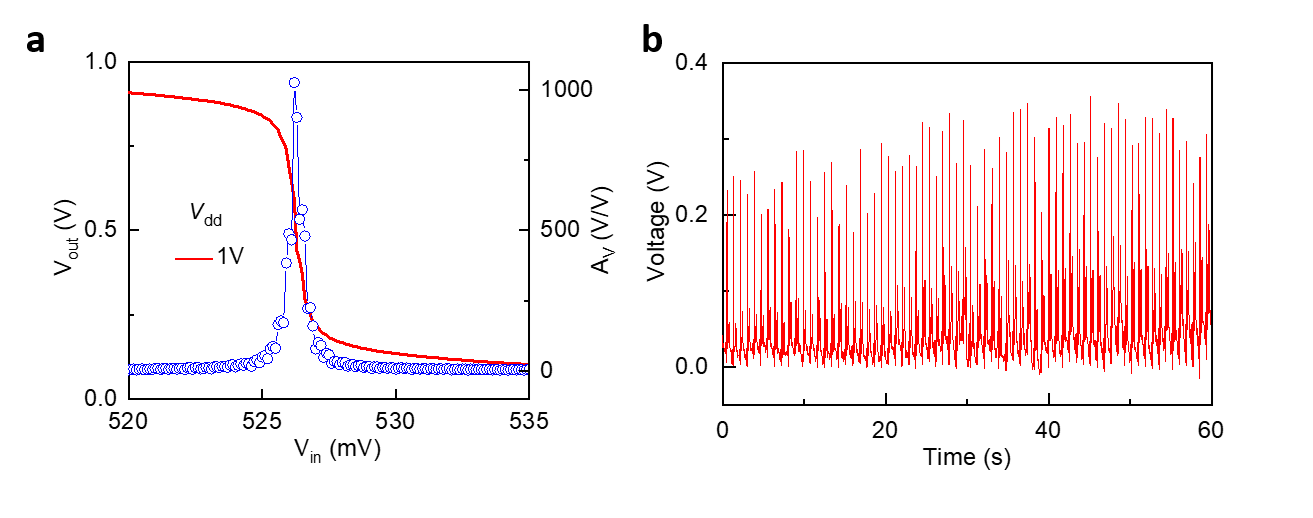
Supplementary Figure 9| Stability of the sub-thermionic OTFTs. a, *I*ds-*V*gs characteristics (*V*ds = -0.1 V) for an OTFT after two-month storage at room-temperature. b, *I*ds-*V*gs characteristics measured at before and after -1.5 V bias stress test. c, Double-sweep *I*ds-*V*gs characteristics (*V*ds = -1 V) at different temperatures of a sub-thermionic OTFT. d, Temperature dependence of *SS* (averaged over one decade of *I*ds) from 200 to 375 K, showing that the *SS* was below the Boltzmann limit in a wide temperature range. Red, forward sweep; blue, reverse sweep.



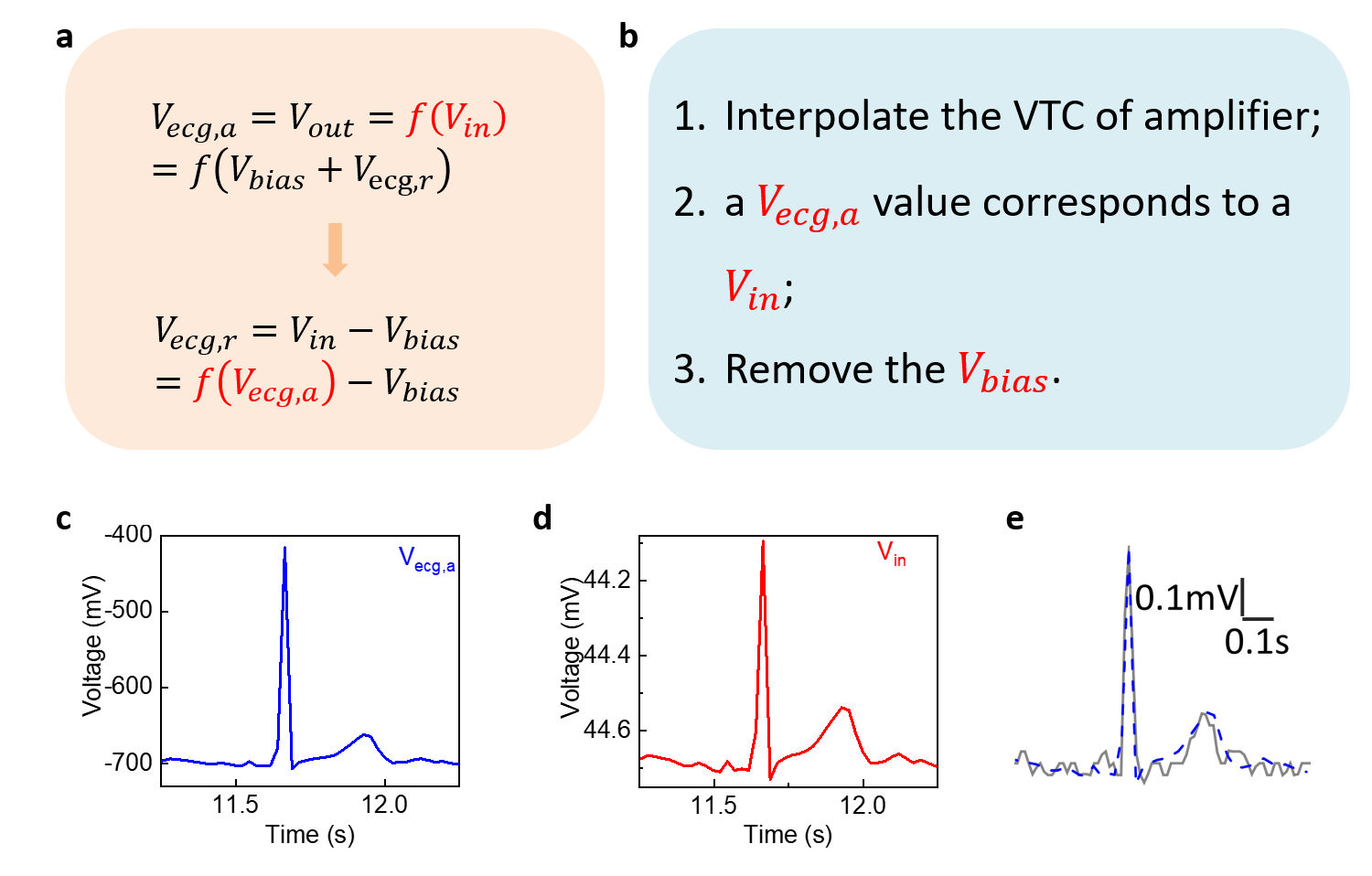
Supplementary Figure 10| Integrated logic gates based on sub-thermionic monolayer OTFT. a,c, Equivalent circuit diagram (left) and optical microscope image (right) of NAND (a) and NOR (c) gates. The dotted boxes indicate organic film areas for transistors. b,d, Output characteristics of NAND (b) and NOR (d) gates.



Supplementary Figure 11| Performance comparison of amplifiers on different dielectrics. a, Transfer characteristics of the load (W/L=180/5) and drive (W/L=90/5) transistors. b, Measured voltage transfer characteristic and power curve of an inverter on HZO substrate. c, Voltage transfer characteristic and d, voltage gain of an inverter on Al2O3 substrate. At *V*dd = -1V, the device on normal Al2O3 dielectric barely shows any gain, which is dramatically different from sub-thermionic OTFTs.



Supplementary Figure 12| Battery-power amplifier module to monitor ECG. a, Voltage transfer characteristic and voltage gain of an inverter powered by a coin battery showing a peak gain greater than 1000 at *V*dd = 1 V. b, ECG monitoring using battery-powered amplifier module.



Supplementary Figure 13| ECG deconvolution. a, The principle of ECG signal deconvolution. b, The specific deconvolution steps. c,d, ECG signal before (c) and after (d) deconvolution. The gain is calculated to be 459. e, Comparison of the deconvoluted ECG signal (blue dashed line) and commercial equipment taken on the same human subject (grey line).

Supplementary Table 1| Contact resistance comparison for OTFTs.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Material | Device structure | Thickness (nm) | Contact technique | *R*c*W*  (Ωcm) | Ref. |
| Rubrene | Bottom-contact | 1000 | Nickel contact | 100 | 8 |
| Pentacene | Bottom-contact | NA | UV/ozone treatment | 80 | 9 |
| C8-BTBT | Bottom-contact | 40 | Doped | 100 | 10 |
| C8-BTBT | Top-contact | 40 | Doped | 200 | 10 |
| C8-BTBT | Top-contact | Monolayer | Graphene contact | 100 | 11 |
| C8-DNBDT-NW | Top-contact | Bilayer | F4-TCNQ/Au | 46.9 | 12 |
| DPH-DNTT | Bottom-contact | 20 | PFBT treatment | 29 | 13 |
| DPH-DNTT | Top-contact | 20 | PFBT treatment | 56 | 13 |
| C10-DNTT | Top-contact | Monolayer | vdW contact | 59.4 | This work |

Supplementary Table 2| Performance comparison of low-voltage OTFT technologies.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Dielectric | Material | Thickness (nm) | *L*/*W* (μm/μm) | *V*dd  (V) | *SS* (mV/dec) | *I*on/*I*off | *g*m (μS μm-1V-1) | Ref. |
| TiO2+SAM | DNTT | 30 | 25/300 | -1 | 77 | 2.1×107 | 2.4×10-2 | 14 |
| AlOX+SAM | DPA | 27 | NA/6.8 | -3 | 66 | 1.1×104 | 3.6×10-3 | 15 |
| AlOX+SAM | DPH-DNTT | 20 | 8/200 | -3 | 62 | 3.2×107 | 5.6×10-2 | 13 |
| AlOX+SAM | DPH-DNTT | 20 | 8/200 | -3 | 68 | 6.7×106 | 5.9×10-2 | 13 |
| AlOX+SAM | DPH-DNTT | NA | 0.6/100 | -3 | NA | 2×107 | 3.7×10-1 | 16 |
| AlOX+SAM | C8-DNBDT-NW | Bilayer | 3/750 | -10 | NA | 4.9×104 | 2.5×10-5 | 12 |
| AlOX+SAM | DNTT | 20 | 50/500 | -2 | NA | 4.1×104 | 3.0×10-3 | 17 |
| HfO2+SAM | Pentacene | 50 | 20/1000 | -3 | 75 | 1.5×105 | 1.8×10-3 | 18 |
| AlOX+PVP | TES-ADT | 50 | 100/1500 | -1 | 85 | 5.2×104 | 2.0×10-3 | 19 |
| PVC | C8-BTBT | 20 | 40/1400 | -3 | 60.2 | 3.5×105 | 4.2×10-5 | 20 |
| Copolymer | Pentacene | 50 | 30/240 | -3 | 220 | 3.7×104 | 5.8×10-4 | 21 |
| PVDF | PTDPPTFT4 | NA | 50/1000 | -5 | 120 | 1.0×103 | 4.1×10-4 | 22 |
| PVP | PTDPPTFT4 | NA | 50/1000 | -5 | 110 | 5.4×104 | 1.3×10-5 | 23 |
| Polymer | C60 | 50 | 200/1000 | 2.5 | 173 | 7.9×102 | 6.1×10-5 | 24 |
| Ba0.7Sr0.3TiO3 | Pentacene | 40 | 100/2000 | -2.5 | 140 | 2.6×103 | 1.3×10-3 | 25 |
| HaLaO | Pentacene | 70 | 80/2000 | -2 | 78 | 1.2×105 | 2.5×10-3 | 26 |
| EDL | C8-BTBT | NA | 100/21000 | -5 | NA | 1.6×101 | 1.1×10-6 | 27 |
| EDL | P(T0T0TT16) | NA | 2.5/1000 | -1 | 70 | 5.8×103 | 1.5×10-2 | 28 |
| HZO | C10-DNTT | Monolayer | 5/180 | -1 | 58.6 | 2.0×108 | 4.1×10-1 | This work |

Supplementary Table 3| Voltage gain comparison of inverters based on different semiconductors.

|  |  |  |  |
| --- | --- | --- | --- |
| Materials | *V*dd (V) | Gain (V/V) | Ref. |
| IGZO | 2 | 220 | 29 |
| ITO | 0.5/2.5 | 178/476 | 30 |
| MoS2 | 10 | 155 | 31 |
| MoS2 | 5 | 60 | 32 |
| WSe2 | 5.5 | 340 | 33 |
| WSe2 | 3 | 12 | 34 |
| MoTe2 | 1/2 | 18/29 | 35 |
| BP | 2 | 13 | 36 |
| MoS2/MoTe2 | 0.5/1 | 7.7/33.3 | 37 |
| C8-BTBT | 2 | 260 | 20 |
| DNTT | 2 | 496 | 17 |
| Pentacene | 1.5/2.5 | 574/478 | 38 |
| DNTT | 3 | 11 | 39 |
| P-Pentacene | 4 | 500 | 40 |
| P-DPH-BTBT | 1 | 180 | 41 |
| CNT | 3 | 290 | 42 |
| CNT | 6 | 25 | 43 |
| CNT | 5 | 16 | 44 |
| CNT | 2 | 30 | 45 |
| C10-DNTT | -1/-2/-3 | 4069/6621/11220 | This work |
| C10-DNTT | -1/-2/-3 | 4825/7690/9762 | This work |

Supplementary Table 4| Performance comparison of amplifiers for detecting biosignals.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| TFT Type | TFT Scale | Circuit topology | Biosignal | Clinical | *V*dd (V) | Battery Power | Gain (V/V) | SNR (dB) | Ref. |
| OFET | 4 | 2-stage inverter | Rat’s ECG | - | 2 | - | ~100 | ~36 | 17 |
| OFET | 4 | 2-stage inverter | Pulse | - | 30 | - | ~10 | - | 46 |
| OFET | 7 | 1-stage differential amplifiers | ECG | - | 4 | - | ~16 | 34 | 47 |
| IGZO TFT | 4 | 1-stage  differential amplifier | HR | - | 10 | - | ~13 | - | 48 |
| Dual Gate IGZO TFT | 5 | 1-stage  differential amplifier | ECG | - | 10 | - | ~60 | - | 49 |
| IGZO TFT | 10 | Differential Amplifier | ECG | - | 10 | - | ~32 | - | 50 |
| CNT-FET | 4 | 1-stage inverter | ECG | - | 10 | - | >60 | - | 51 |
| OFET | 2 | 1-stage inverter | Pulse | - | 1 | Yes | ~900 | 40.4 | This work |
| OFET | 2 | 1-stage inverter | ECG | Yes | 1 | Yes | ~324 | 42 | This work |

Supplementary Movie 1| ECG monitoring by sub-thermionic amplifier and commercial equipment (Prince 180B by Heal Force).

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