

## Supporting Information

### **A metastable small organic molecule for secure memory devices**

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## Experimental Section

### Materials

2,7-Dibromo-9-fluorenone, phenol, 5-aldehyde-2-thiophene boronic acid, 1-pyrenylboronic acid, tetrakis(triphenylphosphine)palladium, methanesulfonic acid, and 2-ethylhexyl bromide were purchased from commercial suppliers (TCI, Sigma-Aldrich, Shanghai Haohong Scientific Co., Ltd). *N,N*-Dimethylaniline, dichloromethane, anhydrous sodium sulfate, ethyl acetate, ethanol, potassium carbonate, and pyridine were purchased from Sinopharm Chemical Reagent. All other reagents and solvents were dried and distilled by standard methods prior to use.

### Characterization

NMR spectra were determined using the Inova 400 MHz FT-NMR spectrometer. UV-vis absorption spectra were obtained from a Shimidazu UV-3600 spectrophotometer. High-resolution mass spectra (HRMS) were obtained from a MicroMass TOF-MS spectrometer (EI). Thermogravimetric analysis (TGA) was conducted on a PETGA-7 thermogravimetric Analyzer with a heating rate of 20 °C min<sup>-1</sup> under nitrogen. X-ray diffraction (XRD) patterns were taken on an X'Pert-Pro MPD X-ray diffractometer. Cyclic voltammetry (CV) measurements were carried out under an argon atmosphere. The film coated on an indium tin oxide (ITO) electrode (working electrode) was scanned at 100 mV s<sup>-1</sup> in a 0.1 M solution of tetrabutylammonium hexafluorophosphate (*n*-Bu<sub>4</sub>NPF<sub>6</sub>) in acetonitrile, with Ag/AgCl (3.8 M KCl) and platinum as the reference and counter electrodes, respectively. A KW-4A-type spin coater produced by the Beijing Institute of Microelectronics was used to prepare active films. The onset oxidation potential ( $E_{ox}^{onset}$ ) of ferrocene was measured to be 0.48 V. Scanning electron microscope (SEM) images were taken on a Hitachi S-4700 scanning electron microscope. Atomic force microscopy (AFM) measurements were performed by using an MFP-3DTM (Digital Instruments/Asylum Research) AFM instrument in tapping mode. Aluminum (Al) electrodes were prepared by a JEOLJEE-400 vacuum deposition apparatus. I-V curves were pictured by using a Keithley 4200-SCS semiconductor parameter analyzer under ambient conditions.

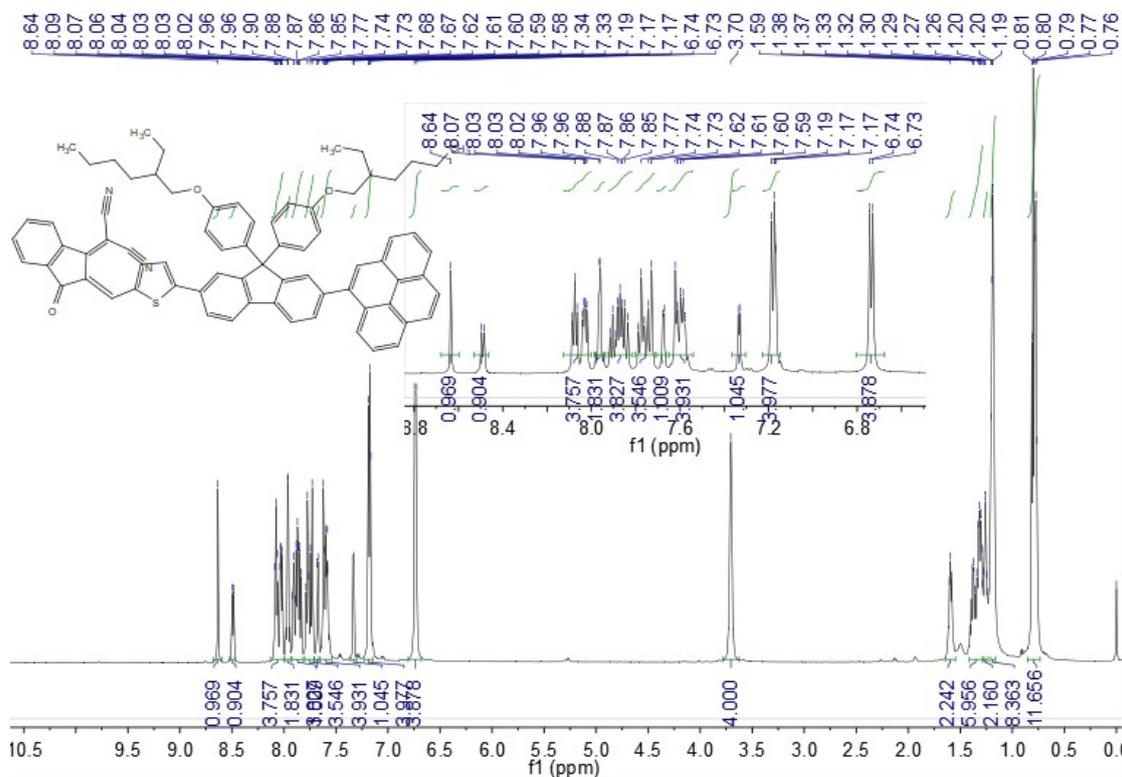
### Fabrication of the memory devices

The ITO glass was carefully precleaned sequentially with deionized water, acetone, and isopropanol by ultrasonic bath for 30 min, and the different thickness of the organic layer (70 nm, 100 nm) was covered on the ITO surface by spin-coating. The above-prepared active film was dried in vacuo for 12 h. A layer of Al, about 120 nm in thickness, was thermally evaporated and deposited onto the organic surface under a pressure of about 5×10<sup>-6</sup> Torr. through a shadow mask to form the top electrode. As a result, an active device area of about 0.0314 mm<sup>2</sup> was obtained.

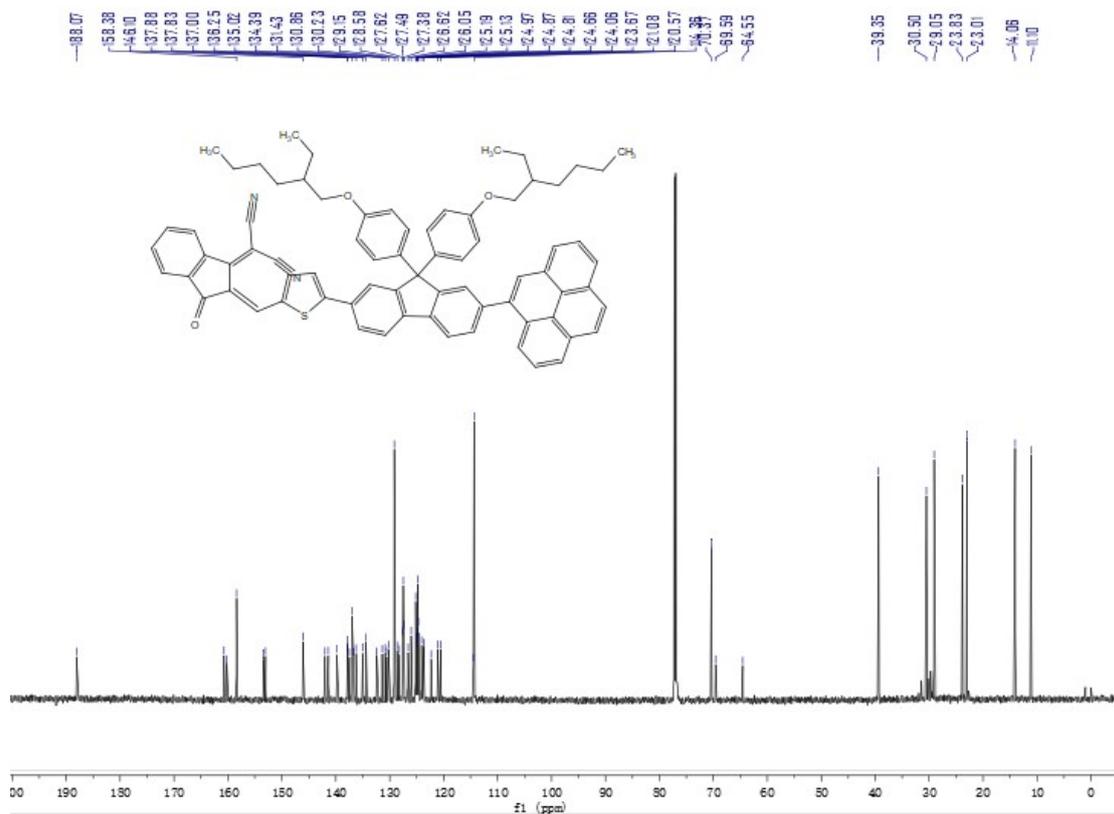
### Synthetic procedures

**Synthesis of PFTCM.** To a 250 mL three-necked round bottom flask were added 5-(9,9-bis(4-((2-ethylhexyl)oxy)phenyl)-7-(pyren-1-yl)-9H-fluoren-2-yl)thiophene-2-carbaldehyde (PFTC, 88.5 mg, 0.1 mmol), dicyanomethylene indeneone (194 mg, 1 mmol), pyridine 0.5 mL and

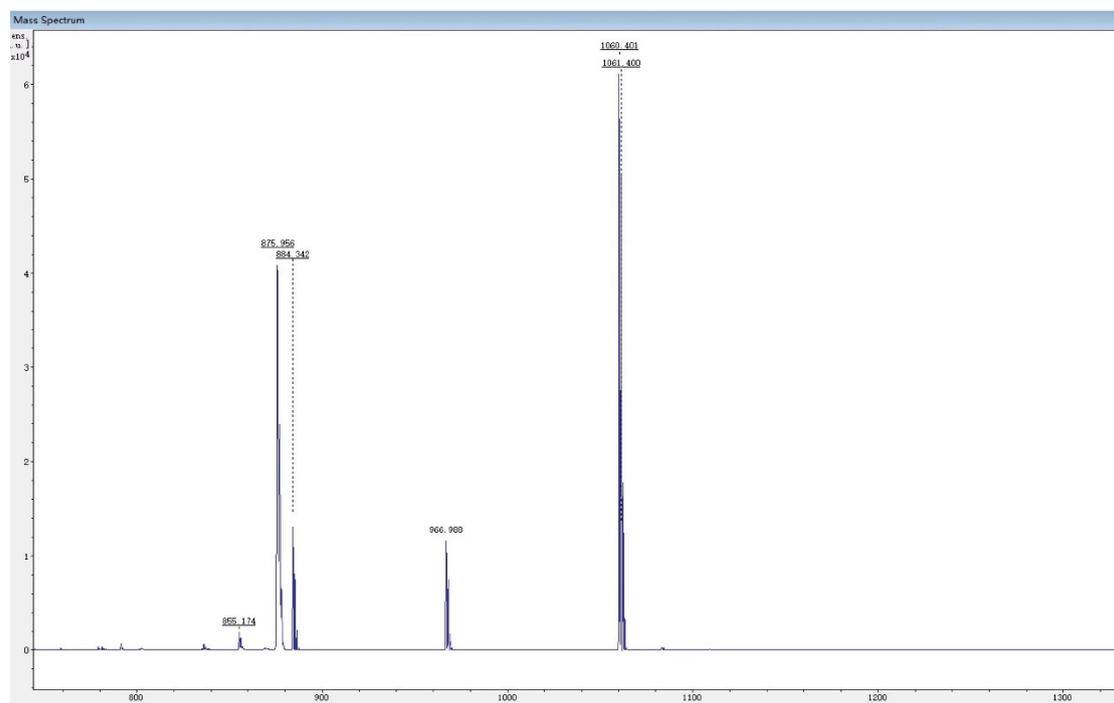
chloroform (50 mL). The mixture solution was stirred and refluxed for 12 h. After cooling down to room temperature, the mixture solution was added into 50 mL ice-water and further extracted with dichloromethane (DCM, 3 × 20 mL). The organic phase was dried over anhydrous Na<sub>2</sub>SO<sub>4</sub> and filtered. After the solvent was concentrated, the residue was purified by column chromatography on silica gel using DCM/petroleum ether (1:5) as an eluant, and an orange solid (Yield: 76 mg, 0.07 mmol, 72%) was obtained. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ = 8.86 (s, 1H), 8.69 (d, *J* = 7.0 Hz, 1H), 8.20 (d, *J* = 7.4 Hz, 2H), 8.15 (d, *J* = 7.8 Hz, 2H), 8.09 (s, 2H), 7.99 (dd, *J* = 20.2, 7.7 Hz, 5H), 7.90 (s, 1H), 7.84 (d, *J* = 11.9 Hz, 2H), 7.76 (s, 2H), 7.70 (s, 2H), 7.50 (s, 1H), 7.26 (s, 4H), 6.81 (d, *J* = 7.7 Hz, 4H), 3.79 (d, *J* = 4.5 Hz, 4H), 1.68 (s, 2H), 1.48 – 1.35 (m, 6H), 1.28 (s, 8H), 0.92 – 0.82 (m, 12H). <sup>13</sup>C NMR (151 MHz, CDCl<sub>3</sub>) δ 188.07, 160.72, 160.21, 158.38, 153.42, 153.03, 146.10, 142.01, 141.40, 139.80, 137.85, 137.44, 137.00, 136.75, 136.25, 135.02, 134.39, 132.39, 131.43, 130.86, 130.61, 130.23, 129.15, 128.58, 128.32, 127.62, 127.43, 126.62, 126.05, 125.04, 124.81, 124.65, 124.06, 123.67, 122.34, 121.08, 120.57, 114.42, 70.37, 39.35, 30.49, 29.05, 23.82, 23.01, 14.05, 11.09. HR-MS, calculation: 1061.3970, found: 1061.4000.



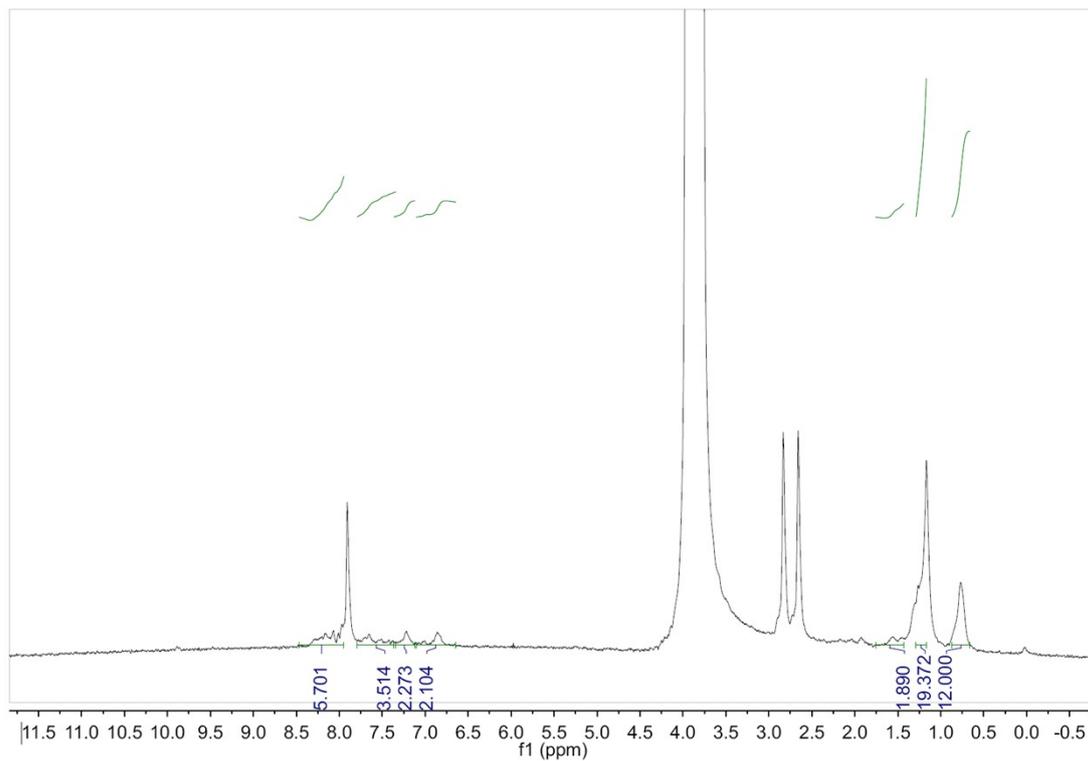
**Fig. S1** The <sup>1</sup>H NMR spectrum of PFTCM.



**Fig. S2** The <sup>13</sup>C NMR spectrum of PFTCM.



**Fig. S3** The HRMS spectrum of PFTCM.



**Fig. S4** The  $^1\text{H}$  NMR spectrum of PFTCM under 365 nm light irradiation for 20 min.