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ELECTRONIC SUPPORTING INFORMATION

Photoresponsive 2D Polymeric Langmuir–Blodgett Films of 2,3,6,7,10,11-Hexaiminotriphenylene

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1. General information

Reagents were commercially available and used without further purification unless otherwise indicated. laser with a wavelength centered at 980 nm were purchased from commercial sources. The HATP polymeric films were prepared by KSV NIMA KN 2002 Langmuir–Blodgett device. Absorption spectra were taken with a CARY 5000 UV-Vis-NIR spectrophotometer. Raman spectrums were captured by Xplora HORIBA. FT-IR absorption spectra were taken with a Shimadzu IRPrestige-21 spectrophotometer. Brewster Angle Microscope (BAM) images were obtained on KSV NIMA Brewster Angle Mic. The X-ray photoelectron spectroscopy (XPS) were obtained on Physical electronics Quantum 2000. Morphology characterizations were obtained on High Resolution Transmission Electron Microscopy JEOL 2100, Transmission Electron Microscopy JEOL 1400, Scanning Electron Microscopy ZEISS SIGMA. Thickness measurements were taken on Atomic Force Microscope (Bruker Multimode V). The electrical measurements were performed at room temperature, using a Keithley semiconductor parameter analyzer (model 4200-SCS). For photocurrent measurements in visible and NIR region, 980 nm laser (200 mW) was used to test power dependence and the wavelength dependence was performed using xenon lamp from 200 nm-1000 nm (CHI). Monochromator could filter out specific wavelengths. An attenuater (THORLABS) was used to damping the laser intensity.



2. The isotherms of repeat experiments for 2D HATP polymeric films

Fig. S1 The isotherms of repeated experiments.

3. Experimental Set-up



Fig. S2 Langmuir–Blodgett device in home-built box.



4. Additional SEM and TEM images for 2D HATP polymeric films

Fig. S3 TEM images of 2D HATP polymeric films.



Fig. S4 SEM images of 2D HATP polymeric films.

5. SEM, AFM, Optical photograph, TEM image of HATP polymeric films.



Fig. S5 (a) Scanning electron microscopy (SEM) images of the films on silicon wafer. (b) AFM image of 2D HATP polymeric films, indicating the thickness of sample of ~ 5.8 nm. (c) Optical photograph of 2D HATP polymeric films on SiO2/Si substrate. (d) High-magnification TEM image of 2D HATP polymeric films on copper grid.

6. SEM-EDX and mapping for 2D HATP polymeric films



Fig. S6 SEM-EDX and mapping for 2D HATP polymeric films transferred to the silicon substrate.



7. Additional AFM images and statistical data of thickness for 2D HATP polymeric films

Fig. S7 AFM images of films from repeated experiments and statistics of film thickness.

8. XPS for 2D HATP polymeric films



Fig. S8 XPS for 2D HATP polymeric films transferred to the silicon substrate.

9. Raman spectrums for HATP molecule and 2D HATP polymeric films



Fig. S9 Comparison of the Raman spectra of HATP monomer and 2D HATP polymeric films with a laser excitation wavelength of 532 nm.

10. Optical images of 2D HATP films device



Fig. S10 The optical images of 2D HATP films device and damaged film.

11. Device characteristics for other 2D HATP polymeric FET



Fig. S11 Transfer characteristics for another two devices. $V_{ds} = -1$ V; channel length (L) = 180 µm; channel width (W) = 1 mm.

12. Statistics of measurements of transport properties of nine devices

Numbers of	Sample								
Sample	1	2	3	4	5	6	7	8	9
conductivity (S cm ⁻¹)	0.69	0.91	0.92	1	1.2	1.71	2	0.31	0.29

Table S1 Statistics of device conductivities

13. I-V curves of 2D HATP polymeric films at drain and source terminal



Fig. S12 I-V curves of 2D HATP polymeric films at drain and source terminal at the same time. ($V_{gs} = 0 V$).

14. UV-Vis-NIR absorption spectra for 2D HATP polymeric films



Fig. S13 UV-Vis-NIR Absorption spectra of 2D HATP polymeric films on SiO₂ substrate.

15. Stability test of device



Fig. S14 Comparison of I-V curves of a device after storage in air for 10 days.