Supporting Information

Molecular Conducting Magnetic Heterostructure

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Experimental Section

Preparation of 2D PANi: Briefly, 20 mg of PANi powder was dissolved in 10 mL of DMSO following by the filtration of undissolved precipitate and then transferred into a petri dish. A piece of oleylamine-molybdenum oxide on a filter paper was added into the petri dish containing PANi DMSO solution. The oleylamine-molybdenum oxide nanosheets serve as the source of oleylamine which is prepared based on the previous literature.[1] The release of oleylamine from oleylamine-molybdenum oxide can trigger the assembly of PANi molecules resulting in the gradual formation of the quasi 2D PANi on the surface of DMSO solution. The solution will turn purple color from the blue color indicating the formation of PANi nanosheet. The as-formed nanosheet was picked up by a substrate (glass, silicon, or patterned ITO electrode) for further characterization and measurement.

Transition metal and HCl doped PANi: For the doping of transition metal, a piece of PANi nanosheet was transferred to 20 mg/mL of $FeCl_3$, $NiCl_2$, or $CoCl_2$ aqueous solution by a glass substrate. For 0.5 h, the transition metal doped PANi nanosheet was picked up by a substrate and annealed at 100 ^oC under vacuum condition. For the HCl doping, a piece of PANi nanosheet was transferred to 1 mol/L of HCl aqueous solution by a glass substrate. After 10 min, 0.5 h, or

2 h, the HCl doped PANi nanosheet was picked up by a substrate and annealed at 100 ^{o}C under vacuum condition.

Characterization: The scanning electron microscopy (SEM) images and element analysis of PANi and doped PANi nanosheets were taken on the Carl Zeiss AURIGA Cross Beam Focused Ion Beam Electron Microscope with an accelerating voltage of 200 KV, and a point resolution of 0.19 nm. The transmission electron microscopy (TEM) images of PANi and doped PANi nanosheets were taken on a high-resolution electron microscope-JEOL JEM 2010. X-ray diffraction experiment was conducted on the Rigaku Ultima IV with operational x-ray tube power of 1.76 kW (40 kV, 44 mA). UV-Vis-NIR absorption spectra was measured by Agilent Cary 7000 Spectrometer. Raman analysis was conducted by Raman microscope (Renishaw InVia). Optical microscopy images were taken by OMRX microscope (A35100U3) equipped with a halogen lamp (6V 30W). Electron paramagnetic resonance (EPR) measurement of $FeCl_3$-doped PANi was characterized by EMX 390 EPR Spectrometer at 293 K with a microwave frequency of 9.838 GHz and a microwave power of 1.58 mW. The electrical performance (I-V curves) was tested by Keithley 2450 sourcemeter. Resistance-temperature curve of HCl-doped nanosheet was taken in a high vacuum container equipped with a Keithley 2400 sourcemeter and a LakeShore temperature controller. Low temperature magnetic properties measurements were conducted on a Physical Properties Measurement System (PPMS, Quantum Design) equipped with a Vibrating Sample Magnetometer (VSM). Magnetoconductance measurement was measured by Quantum Design Physical Property Measurement System (PPMS) EverCool-II. Atomic Force Microscopy mapping of bilayer structure was taken by Bruker Dimension Icon Atomic Force Microscope System with ScanAsyst.

Reference

[1] Q. Huang, S. Hu, J. Zhuang and X. Wang, Chem. - Eur. J., 2012, 18, 15283

Figure S1. Photographs of assembled PANi with increased of the amount of oleyamine after 2 h. Increased oleyamine was achieved by increasing the amount of oleylamine-molybdenum oxide nanosheets on filter paper. Detailly, 0.75 g of ammonium molybdate was dissolved in 37.5 mL of H_2O. Then, 15 mL, 0.3 mol L^{-1} of HCl solution was added following by addition of a mixture of 15 mL of hexane and 3 g of oleylamine. Under stirring, the solution become a milky suspension which was then transferred into a Teflon-lined autoclave with stainless steel shell for the hydrothermal reaction under 180 °C for 5 h. The as-synthesized molybdenum oxide nanosheets with oleyamine as surfactant were washed using hexane and ethanol and then redispersed in 20 mL of hexane. Then, 0.1 mL, 0.25 mL, 0.5 mL, and 1 mL of oleylamine-molybdenum dipsersion in hextane were respectively filtered onto the filter paper for the assembly of PANi nanosheets from left to right in the above figure.

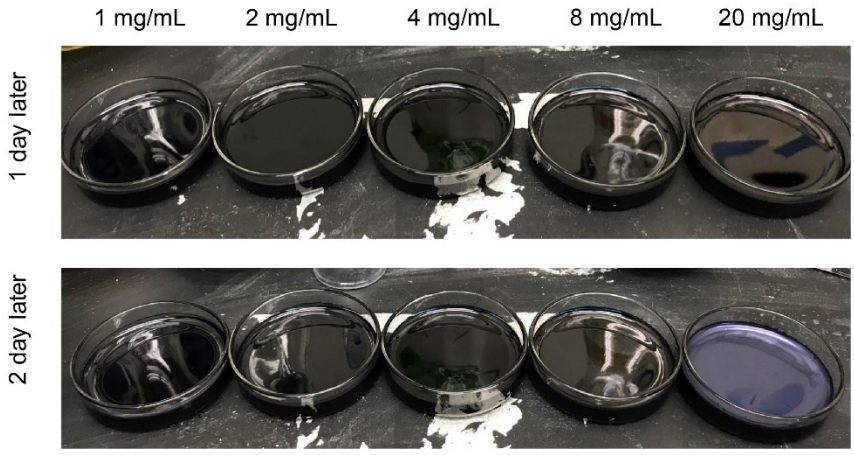

Figure S2. Photographs of the formation of PANi on the DMSO surface without oleyamine. In this case, only the DMSO solution containing 20 mg/mL of PANi shows the self assembly of PANi by observation.

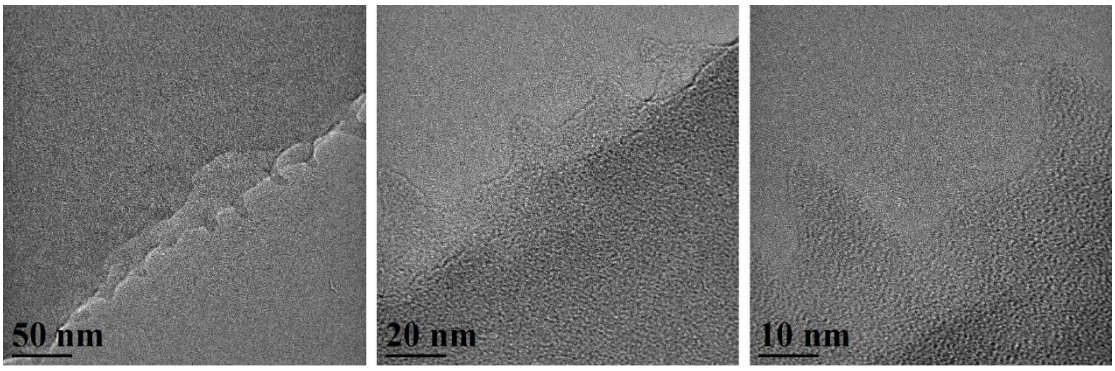

Figure S3. TEM images of PANi nanosheet.

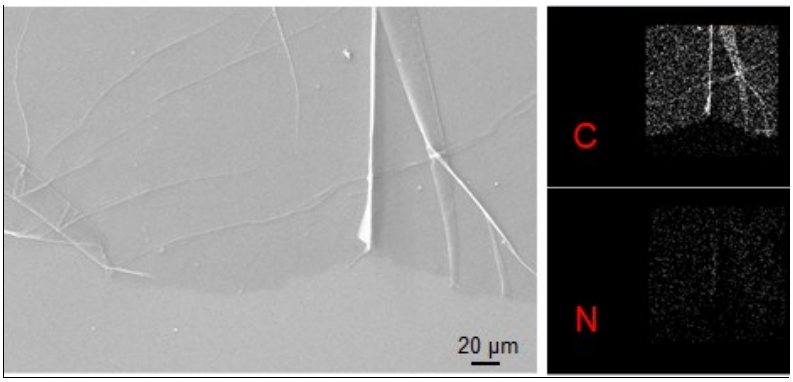

Figure S4. SEM image and EDS mapping of HPANi nanosheet.

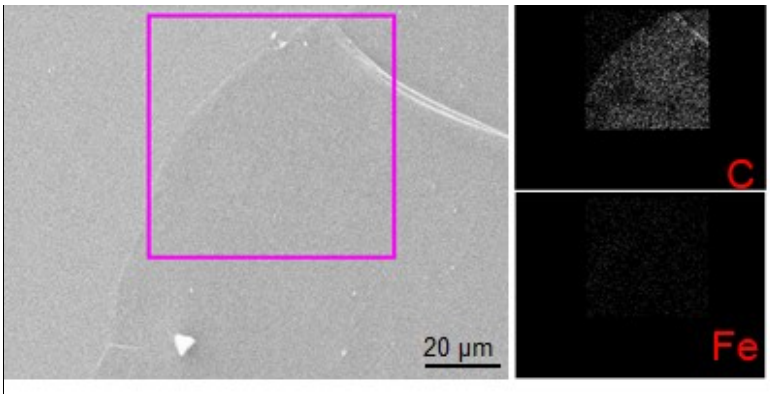

Figure S5. SEM image and EDS mapping of FePANi nanosheet.

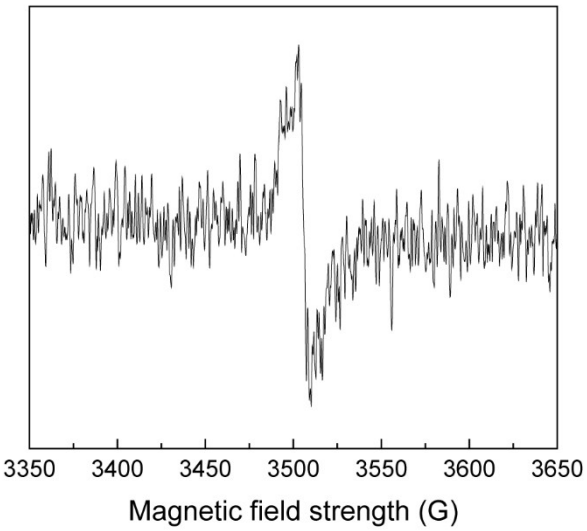

Figure S6. EPR spectrum of FePANi nanosheet. The g factor was calculated to 2.0054, which is close to the value of the free electron (2.0023), indicating the presence of unpaired electrons in the FePANi nanosheets, which is the prerequisite of ferromagnetism.

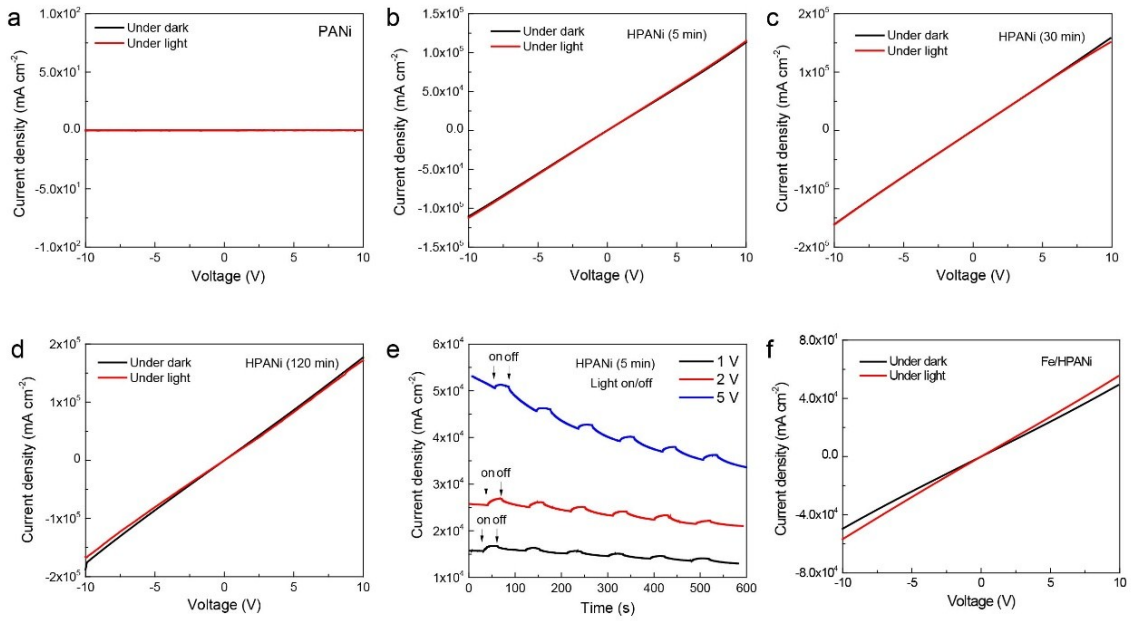

Figure S7. Current-voltage curves of a) PANi, b) HPANi (5-min doping), c) HPANi (30-min doping), d) HPANi (120-min doping) under dark and light supplied by a solar simulator with a light intensity of 5 W/cm^2. e) Current-time curves with light on and off. f) Current-voltage curves of FePANi/HPANi heterostructure under dark and light condition.